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Assessment of pilot-scale acid washing of soil contaminated with As, Zn and Ni using the BCR three-step sequential extraction

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Abstract

This study performed pilot-scale washing of soil contaminated with both oxyanion and cations as a recalcitrant remediation case due to their different chemical behavior. The soil contaminated with As, Zn and Ni, partially recalcitrant due to their strong binding properties, was obtained near a closed iron/serpentine mining area. This study monitored the variation of chemical speciation of As, Zn and Ni for acid solutions and particle size fraction using the BCR sequential extraction and evaluated the optimal condition of physical separation of highly contaminated fine particles for enhanced washing. H_2SO_4 and H_3PO_4, including competitive oxyanions, enhanced removal of As with the simultaneous extraction of Zn and Ni. Less nickel from the residual fraction in coarse particles was extracted than As and Zn due to the recalcitrant serpentine. Fe/Mn oxide, organic/sulfides and residual fractions in fine particles were enriched with contaminants due to the high surface areas and recalcitrant minerals. The chemical extraction of As was also restricted in the fine particles, whereas the chemical extraction of Zn and Ni was determined by the residual form of various particle size fractions. Further extraction was limited in the exchangeable and residual fractions and retained a gradual extraction from Fe/Mn oxide and organic/sulfides fractions, which indicated an instant detachment from the easily bound fraction. Correspondingly, extraction from the acid-attackable fraction was related to the exchangeable Fe/Mn oxide and organic/sulfides fractions. Due to the limitation of chemical extraction, the physical separation of fine particles could enhance the effectiveness of acid washing. In addition, the chemical properties of the soil were affected by strong acid washing. The treated soil then needed to be regenerated.

Keywords: Arsenic; Zinc; Nickel; Acid washing; BCR sequential extraction

1. Introduction

Contamination by As (arsenic) and heavy metals is problematic in the geochemical environment around metal mining areas due to arsenic-related chemical spills in agricultural and industrial areas [1,2]. Arsenic and heavy metals have been also contaminants in metallic ores and industrial effluents in Korea [3–6]. Exposure to arsenic and some metals carries risks for humans, such as cancer. The remediation of soil contaminated with As and heavy metals is an important problem for many countries [2]. Soil contaminated with As and heavy metals have been physically, chemically and biologically treated. Physical methods include separation, carbon adsorption, vitrification and incineration. Chemical methods such as solidification/stabilization, encapsulation and washing remove metals or immobilize them by reducing bioavailability. Biological methods use plants or microbes for the removal of metals. The simultaneous removal of As and heavy metals is strongly dependent on chemical speciation with respect to the redox potential and pH [1,7]. Specifically, the solubility of most cationic metals decreases as the solu-

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tion pH increases. However, the solubility of oxyanions such as As, Se, P and Mo increases as pH increases [8]. It is not easy for both oxyanion and cation to find optimal conditions for leaching or immobilization.

Soil washing technology has been applied to the decontamination of heavy metal-contaminated soil such as EDTA and DTPA extraction of Zn [9], EDTA extraction of Pb [10,11] and acid washing of heavy metals [12]. Soil washing of As has been also treated with sodium hydroxide extraction in an alkaline environment [13], phosphate extraction [14] and acid washing [15]. There have been only a few cases described about the simultaneous removal of As and heavy metals. For the simultaneous removal of As and heavy metals, an optimal remedial condition should be set up. This study provides an example of an optimal site-specific study. Two predominant operating factors in soil washing are the chemical extraction of contaminants and the physical size separation of fine particles. Soil remediation is a site-specific process, and, in particular, the chemical speciation of As and heavy metals should be significantly monitored for optimal extraction due to the nature of their binding strengths. Chemical fraction of metals with weak bonding strength can be mobilized and available for uptake. However, metals that have bonded strongly with soil minerals are less exchangeable. It is important to understand the chemical forms of metals for remedial work. Many researchers have suggested modified sequential extraction schemes for selective leaching of particular chemical forms of elements from sediment, soil or waste [6,16–19]. Further, sequential extraction can assess the effectiveness of the remedial processes such as electrokinetic removal of heavy metals [20,21], the application of till cover of mine tailings [22] and the stabilization of As [23]. Particularly, a group in a Community Bureau of Reference (BCR) project proposed a three-step extraction procedure [18,24]. The BCR scheme is now widely used in the analysis of sediment, soil, sludge and mine waste. Therefore, the BCR method can be applied to monitor chemical forms of metals during soil washing in order to evaluate the proper extraction efficiency of contaminants. The physical size separation, a factor that affects enhanced soil washing, can control the levels of contaminants in the treated soil. The fine particles have a high surface area for the adsorption of contaminants; therefore, the concentrations of contaminants are extremely high. However, the extraction of metals in the fine particles is very poor, less movable and available for uptake in the ecosystem. If the fine particles are properly separated, the overall effectiveness of soil washing is enhanced.

In this study, the pilot-scale washing of soil contaminated with both As and heavy metals was performed as a recalcitrant case. The objectives of this study are to find the optimal conditions of pilot-scale acid washing for the simultaneous removal of As and heavy metals and to evaluate the characteristics of soil contaminants using the sequential extraction method during soil washing. Specifically, we characterized the soil properties for soil washing in the contaminated area, the effect of the acid solution type and the effect of the reaction time for acid washing. Then, we monitored the variation of chemical fractions of As, Zn and Ni with respect to both the acid solutions and particle sizes during the acid washing, and determined the optimal size for physical separation of fine particles as a residual form of contaminant.

2. Materials and methods

2.1. Site description and sampling

The Dalcheon iron mining area is located in the southeastern part of Seoul, Korea, approximately 450 km (Fig. 1). The study area will be a hilly district on a plain with a res-

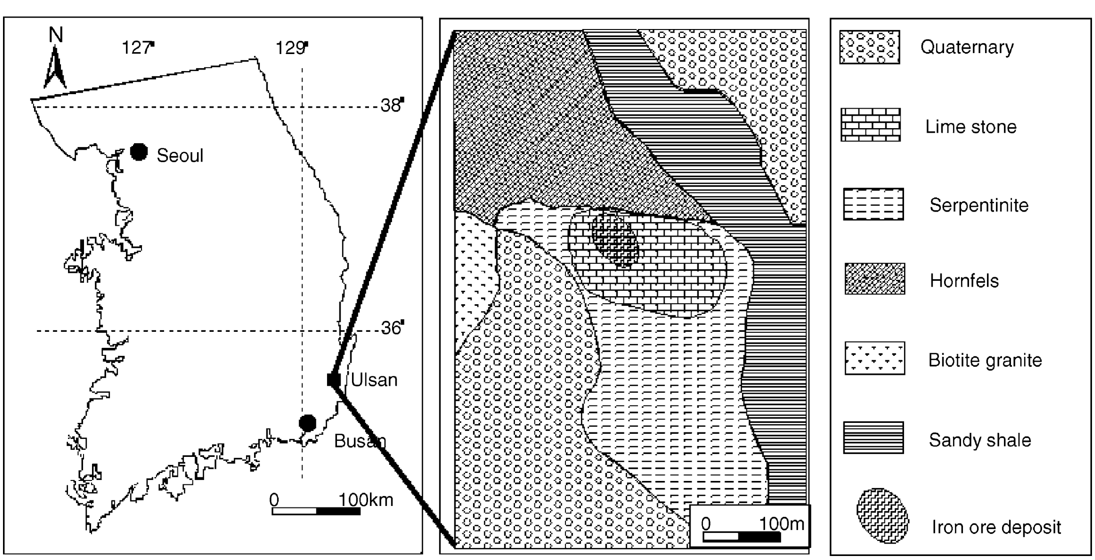

Fig. 1. Study site and geological map.