

Copper leaching from a sandy soil: Mechanism and parameters affecting EDTA extraction

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Abstract

A series of 24 h batch tests of copper extraction from a sandy soil were performed by washing the soil with aqueous solution of ethylenediaminetetraacetic acid, EDTA. EDTA versus copper molar ratio were in the range between 1 (equimolar tests) and 13.3. The tests were performed at three selected liquid/solid ratio, 5, 12.5 and 25. Results show that decreasing the pH of the washing solution an higher copper extraction yield was achieved: an almost complete copper extraction was achieved after 23 h of mixing at a L/S = 5, and after 5 h of mixing at an L/S = 12.5. The mechanism of copper extraction was found to involve two sequential processes: the former dissolution of metal salts, that lead to an initial high concentration of both copper and some competitive cations (essentially Ca^{2+}), and the following EDTA exchange reaction between calcium and copper complexes, which corresponded an increase of pH in the washing solution. A negligible extraction of Fe^{3+} was also observed at the investigated operative conditions.

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1. Introduction

In this paper, an experimentation devoted to copper (Cu) extraction from a contaminated soil is presented. An artificially contaminated sandy soil was used. The extraction was performed by washing the soil with an aqueous solution of ethylenediaminetetraacetic acid (EDTA, EDTA-H_2) [1].

The objective of the work was to investigate the mechanism of copper extraction and to evaluate the influence of the main parameters affecting the washing process, such as the EDTA/metal molar ratio, and the pH and EDTA concentration in the extraction solution. A special attention was devoted to the competitive effect on copper chelation of the other exchangeable cations present in the soil. For the tested soil the final goal was to remove enough copper to achieve a residual copper level in the soil suitable to a civil (120 mg Cu/kg of

soil) or at least an industrial (600 mg Cu/kg of soil) reuse of the soil, according to Italian Environmental Regulation [2].

In the ex situ soil washing [3,4] technology undesirable contaminants in soils are removed by dissolving or suspending them in a washing solution. In the case of heavy metal contaminated soil, the washing solution is generally an aqueous solution of a chelating agent. In the remediation of copper contaminated soil, the effectiveness of EDTA has been widely demonstrated as a chelating agent in washing treatment [5–8].

In comparison with other chelating agents, EDTA presents the following advantages: a low degree of biodegradability in groundwater [4] and soil [1,9,10] and, moreover, a high level of complexing capacity with respect to heavy metals [5,9,11]. Conversely, the low selectivity of EDTA causes a great consumption of this reagent due to the potential chelation of all the exchangeable cations present in soil, such as Ca^{2+} , Fe^{3+} [12].

A substantial excess of EDTA is so generally adopted, thus increasing the cost of the whole remediation treatment.

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Table 1
Composition of the soil used in the experiment (ASTM classification)

Component	Gravel	Coarse sand	Medium and fine sand	Silt and clay
Particle size (mm)	>2	0.425–2	0.075–0.425	<0.002
vol.%	–	9	75	16

Several papers deal with the extraction of heavy metals, and copper in particular, by soil washing but the aim of most of them was to compare the effectiveness of different chelating agents (EDTA, citric acid) towards selected metal extraction or to optimise the solid/liquid ratio to achieve a stated metal extraction yield [9,10,13,14].

Only few papers focus their attention to the mechanism of metal extraction, since a lot of chemical reactions and physical processes are involved [15].

In this paper, a simple model is presented to illustrate the mechanism of the leaching process from a sandy soil: the initial dissolution of copper precipitates was followed by the chelation by EDTA, that, due to its non-selective nature, binded progressively all the dissolved or exchangeable cations until the equilibrium conditions did not occur.

2. Materials and methods

2.1. Soil characterisation

The soil used in the experiments was a volcanic loamy sand collected in a cave near Bracciano, Rome, Italy. It was chosen as representative of a typical sandy soil in central southern Italy.

The soil was initially passed through a 2 mm sieve: the resulting soil composition is given in Table 1. The soil was mixed for 24 h in a Hobart-type mixer at 120 rpm.

After chemically and physically characterizing the soil, 1 kg samples were placed in plastic containers prior to artificial contamination.

Table 2 shows the characteristics of the soil used, determined according to Italian regulation [16]. pH was measured after mixing 10 g of soil samples with 25 ml of a 0.01 M solution of CaCl₂.

The total porosity was determined using 100 g of air-dried soil [17]. The sample was weighed: then a known amount of water was added until saturation was obtained. The total porosity (f) was determined from:

$$f = \frac{V}{V + V_s} \quad (1)$$

where V is the volume of water added and V_s , the volume of the dried soil particles [17], calculated as:

$$V_s = \frac{M}{\rho_s} \quad (2)$$

where M is the mass of the dry soil (g) and ρ_s (g/cm³) is soil particles density.

Table 2
Characteristics of the soil used

Parameter	Value
pH	8.2
Organic carbon (%)	0.9
Water permeability (cm/s)	3.18×10^{-2}
Total porosity (%)	46
Moisture content (g/kg)	24.5
Bulk density (g/cm ³)	1.25
Soil particle density (g/cm ³)	2.3
Pore volume (ml)	32
CaCO ₃ (g/kg)	139
Conducibility (mS)	0.217
Metals (mg/kg)	
P	90
Fe	12.2
Mn	3.8
Cu	2.2
Zn	5.2
Ca	1550
Mg	230
K	260
Na	120
Cationic exchange capacity (meq/100 g)	
Total	10.85
Calcium	7.75
Magnesium	1.92
Potassium	0.66
Sodium	0.52

The pore volume of the soil (PV) was calculated using:

$$PV = \frac{f}{100} \times V_c \quad (3)$$

where V_c is the column volume.

The soil permeability was determined according to the ASTM D2434 standard test (Liu and Evett [18]).

2.2. Soil contamination

Soil contamination was carried out by mixing for 48 h at 120 rpm in a Hobart-type mixer 1 kg of soil and 0.51 of a saturated aqueous solution of dihydrate copper chloride, CuCl₂·2H₂O. After mixing, the samples were air dried by exposure to ambient air for 30 days and stored in plastic containers.

The contamination was carried out so as to obtain a Cu concentration of at least twice the Italian regulation limit for an industrial site.

Subsequent to preliminary acid digestion, a Philips PU 9200 atomic absorption spectrophotometer was used to determine the metal content of six 1 g samples of each contaminated soil. The average Cu concentration was 1230 mg Cu/kg with a standard deviation of ± 98.5 mg.

Samples (100 g) were prepared from the contaminated soil and stored in plastic containers.

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