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Removal of metals and phosphorus recovery from urban anaerobically digested sludge by electro-Fenton treatment



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HIGHLIGHTS

- First application of electro-Fenton (EF) process as urban sludge washing technique
- Acidification and conventional Fenton treatment allowed the leaching of Cd, Cu and Zn.
- Total removal of Cu and Zn using EF with boron-doped diamond or RuO₂based anode.
- P recovery of about 74%–79% in the solid phase achieved by all Fenton-based treatments
- •OH and active chlorine oxidative action on metal-organic complexes promotes leaching.

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G R A P H I C A L A B S T R A C T



ABSTRACT

To our knowledge, this work presents the first application of electro-Fenton (EF) process to sludge washing. Suspensions of anaerobically digested sludge (0.50 wt%) from a municipal wastewater treatment facility were electrolyzed with addition of Na₂SO₄ and Fe²⁺ at pH 3.0, using a stirred tank reactor with a boron-doped diamond (BDD) or RuO₂-based anode and an air-diffusion cathode that produced H₂O₂. The effect of the sludge content in suspensions and applied current density (*j*) was examined. High quantities of Cr, Pb, Cd, Zn, Fe and P were leached at pH 3.0, whereas Cu showed the opposite trend. Aeration only enhanced Pb and Zn leaching, whereas the use of Fenton's reagent with 15 mM H₂O₂ solubilized 16.0% Cr, 23.0% P, 42.6% Fe and 56.0% Pb, with total leaching of Cd, Cu and Zn. EF with BDD anode at high *j* caused total precipitation of Cr, Pb and Fe, 40% Cd leaching and total solubilization of Cu and Zn. The RuO₂-based anode enhanced the entrapment of Cr, Fe and P in the solid fraction of the sludge, but promoted a high transport of Cd, Cu and Zn to the liquid phase. P recovery was about 74%–79% in all EF treatments. The soluble organic carbon increased in most cases except for EF with BDD, where it decreased markedly, in agreement with the high oxidation power of this anode. The sludge dewaterability was largely improved in all treatments, attaining up to 97%, consistent with the scission of many extracellular polymeric components.

1. Introduction

Municipal wastewater treatment facilities (WWTFs) produce large amounts of anaerobically digested sludge from mixed urban and industrial effluents, needing proper management or valorization before disposal (Zhu et al., 2013; Krüger and Adam, 2015). Management of a

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Table 1

Total solid (g of dry sludge per kg of raw sludge) and selected elements concentrations (mg per kg of dry sludge (TS)) obtained for the samples tested in this work.

Sample	TS	Cr	Pb	Cd	Cu	Zn	Fe	Р	С
	$(g kg^{-1})$	$(mg (kg TS)^{-1})$							
S1	38.1 ± 2.3	41 ± 1	18 ± 1	1.4 ± 0.1	313 ± 10	417 ± 12	$\textbf{25,628} \pm \textbf{373}$	$32,661 \pm 247$	$13{,}992\pm195$
S2	32.2 ± 0.2	103 ± 4	48 ± 3	1.9 ± 0.1	517 ± 19	561 ± 17	$67,533 \pm 790$	$41,010 \pm 352$	$11,782 \pm 160$
S3	31.5 ± 0.5	104 ± 2	56 ± 2	1.4 ± 0.1	513 ± 13	616 ± 16	$71,\!120\pm860$	$\textbf{36,836} \pm \textbf{326}$	9455 ± 120

large portion of the produced sludge is based on incineration or landfilling, but the real challenge is to ensure conditioning to serve as raw material for fertilizers, thus giving added value to its high content in nutrients like nitrogen and phosphorus (Ito et al., 2013; Zhang et al., 2017). However, anaerobically digested sludge contains hazardous materials such as pharmaceuticals, endocrine disruptors and metals as a result of ineffective wastewater treatment, thereby requiring some additional step to minimize their content before agricultural usage (Ito et al., 2013; Fontmorin and Sillanpää, 2017). In the case of metals, it has been shown that their removal depends on the chemical structure of the substances contained in the solid fraction of the sludge as well as on the nature of its matrix (Tyagi et al., 1997; Fuentes et al., 2004; Krüger and Adam, 2015; Fang et al., 2016).

Different methods have been utilized to wash urban anaerobically digested sludge regarding metal content. They include acidification (Yoshizaki and Tomida, 2000; Stylianou et al., 2007; Deng et al., 2009; Kuan et al., 2010; Ottosen et al., 2013), chemical treatment (Ren et al., 2015; Wu et al., 2015), hydrolysis (Suárez-Iglesias et al., 2017), wet oxidation (Suárez-Iglesias et al., 2017), bioleaching (Yoshizaki and Tomida, 2000; Zhu et al., 2013), electrodialysis (Ebbers et al., 2015) and electrokinetics (Tang et al., 2017, 2018). Furthermore, advanced oxidation processes (AOPs) such as chemical Fenton-based processes have





Fig. 1. (a) SEM image $(300 \times)$ and (b) EDX analysis of the dry sludge sample S1.

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