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Sequential electrodialytic recovery of phosphorus from low-temperature gasification ashes of chemically precipitated sewage sludge

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ABSTRACT

Phosphorus recycling from secondary materials like sewage sludge ashes offers an alternative to mining of phosphates from primary resources and a mean to counteract the current phosphorous rock depletion concern. A separation of P from the bulk ash is normally required, due to its low plant availability and the presence of heavy metals. Previously, more than 80% of P was recovered from incineration sewage sludge ashes using a two-compartment electrodialytic cell. In contrast, the recovery was below 30% for ashes from low-temperature gasification using the same setup. The low recovery was due to a high presence of Al- and Fe(III)-P bindings. In the present study, an electrodialytic process combining sequentially a pair of two-compartment cells allowed a recovery of up to 70% of phosphorus from these ashes. The use of a second cell, where the ash was suspended in an alkaline solution, allowed the P solubilisation from aluminium and ferric phosphates. In addition, P was separated from most metals as they became insoluble under the prevailing chemical environment. The obtained ratio of Al, Fe, Mg and most heavy metals to P was comparable to wet process phosphoric acid. Therefore, this sequential process was found to be suitable to recycle P and potentially use it in the production of common fertilizers like diammonium phosphate.

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1. Introduction

Europe is dependent on phosphate rock imports (van Dijk et al., 2016), the reserves of which are estimated to be exhausted within 50–400 years (van Dijk et al., 2016; Cordell et al., 2009; Dawson and Hilton, 2011). The European Commission included this mineral in the list of 20 critical raw materials in 2014. Moreover, mineral P-fertilizers are the main contributors of toxic elements accumulation such as cadmium (McBride and Spiers, 2001) and uranium (Taylor, 2007) in agricultural soils. For these reasons, P recycling from secondary resources, like sewage sludge, has gained increased attention among researchers, private companies and public institutions over the last decade (Sartorius et al., 2012; Gorazda et al., 2013; Schoumans et al., 2015; Herzel, 2015). Around 16% of phosphate rock import can be potentially substituted with unrecycled P from sewage sludge in Europe (van Dijk et al., 2016). In order to promote P recycling, some countries e.g. Denmark have set goals for P recovery (Denmark without waste, 2013), and others e.g. Ger-

many, Switzerland and Sweden currently have proposals to make it obligatory. The content of heavy metals as well as potential unwanted organic pollutants and pathogens are of major concerns in respect to sewage sludge's direct use on agricultural fields (Schoumans et al., 2015; Withers and et al., 2015). Another difficulty is the limited plant-availability of P when sewage sludge is precipitated with Al and Fe salts in the wastewater treatment plant to avoid eutrophication of natural water system recipients, a practice which is common in many countries (Gorazda et al., 2013; Paul et al., 2001; Donatello et al., 2010; Rodriguez-Garcia et al., 2011; Kruger et al., 2014).

Thermal processes such as incineration (Donatello and Cheeseman, 2013) and low-temperature gasification (Thomsen et al., 2015) can lead to the removal of organic contaminants from sewage sludge, but additional treatments of the resulting sewage sludge ashes (SSA) are required to separate P from heavy metals and ensure its bioavailability (Gorazda et al., 2013; Schoumans et al., 2015; Franz, 2008; Parés Viader et al., 2015). Electrodialysis (ED) has been studied as a technique to extract P from different SSA using 2-compartment cells (Parés Viader et al., 2015; Ebberts et al., 2015). In this setup, SSA is suspended in water in the anode com-

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partment, which is separated from the cathode compartment by a cation exchange membrane. When applying the electric field the cations are transported from the anolyte to the catholyte through the membrane. Protons are concurrently produced from water electrolysis at the anode, which increases the solubilisation of the different elements present in the SSA (Parés Viader et al., 2015; Ebberts et al., 2015). Thus, the applied electric field results in acidic leaching combined with cation migration into the cathode compartment. Large recoveries of P (higher than 80%) have been achieved for incineration SSA (Ebberts et al., 2015) and biologically precipitated SSA from low-temperature gasification (Parés Viader et al., 2015). On the contrary, less than 26% of P was recovered from chemically precipitated SSA from the same gasification unit. The reason for the low recovery was found to be the influence of Fe (III) on P-solubility and the formation of positively charged Fe-P complexes in acidic medium (Parés Viader et al., 2015).

Other processes than ED has been investigation for the recovery of P from SSA and the majority can be grouped into either:

- (a) Acid extraction of P from SSA with H_2SO_4 , HCl, HNO_3 and/or H_3PO_4 , followed by further steps to separate P from the leached impurities like heavy metals by means of sulphide precipitation (Franz, 2008), solvent extraction (Dittrich et al., 2009) and/or ion exchange columns (Donatello et al., 2010; Franz, 2008; Lehmkuhl and Lebek, 2015a; Takhim, 2015), as well as through the chemical precipitation of P as Ca-phosphates (Herzel, 2015; Lehmkuhl and Lebek, 2015a, 2014, 2015b). The biggest industrial agreement to recycle P from SSA until now was signed in the beginning of 2015 between the Belgian company Ecophos and the Dutch companies HVC and SNB, which includes the construction of a plant to treat 50–60,000 tonnes of incineration SSA yearly using a method belonging to this group.
- (b) Thermal removal of heavy metals at high temperatures (above 1400 °C) Herzel, 2015; Schönberg et al., 2014, which also allows the separation of the main metals like Fe; or at around 1000 °C in combination with chemicals like calcium or magnesium chloride (Adam et al., 2009), Na- and K-salts and bases (Herzel, 2015; Krüger et al., 2016; Stemann et al., 2015), which at the same time increases the plant availability of P in the ashes.

However, these researches have focused on chemically precipitated incineration SSA, where Fe(III) is not controlling P solubility (Cohen, 2009; Gorazda et al., 2012), which is on the contrary to gasification SSA. An alkaline extraction step could potentially dissolve Fe(III)-phosphates (Sano et al., 2012; Wilfert et al., 2015) existing in gasification SSA, with the advantage of preventing the formation of complexes like $\text{Fe}(\text{H}_2\text{PO}_4)^{2+}$ or $\text{Fe}(\text{HPO}_4)^+$ which complicate P separation in ion exchange processes and ED (Parés Viader et al., 2015; Ottosen et al., 2014). By such treatment, alkaline-insoluble Ca-phosphates (Stumm and Morgan, 1996) would, however, not be dissolved. Instead, a common strategy to leach P from materials containing both Ca-P and Al/Fe(III)-P bonds, previously used for Al-precipitated SSA (Petzet et al., 2012, 2011), soils (Benzing and Rickardson, 2005), sediments (Ribeiro et al., 2008), and municipal solid waste fly ashes (Kalmykova and Fedje, 2013), is to solubilise the different Ca-bearing phases in an acidic step, and the Al/Fe(III)-P bindings in an alkaline step. Nevertheless, the order of execution requires an investigation. Starting with an acidic extraction can lead to P re-precipitation as Al-P (Petzet et al., 2012, 2011), and probably Fe(III)-P, which increases the hydroxide demand for their dissolution in the subsequent alkaline step. Starting with an alkaline extraction, P could re-precipitate as Ca-P or adsorb to Ca compounds (Petzet et al., 2011; Benzing and Rickardson, 2005; Chen et al., 2007), which increases the proton

demand for P dissolution in the following acidic step. In order to assess which combination is best to recover P, both possible two-stage chemical processes are compared in the present study.

Recently, a patent concerning an innovative sequential ED process (Fig. 1) has been filed with the European Patent application number 15187370.0. The new method combines two existing 2-compartment cell configurations, one with a cation exchange membrane (Parés Viader et al., 2015; Ebberts et al., 2015) and another one with an anion exchange membrane (Touaibia et al., 1996), into a sequential process. The present research focuses on achieving a higher recovery of P from low-temperature gasification SSA, which has been chemically precipitated, using a sequential ED process in comparison to a single step with a 2-compartment cell with a cation exchange membrane.

2. Experimental

2.1. Experimental SSA

The investigated SSA were the same as in previous published work (Parés Viader et al., 2015), where the ashes were collected from the second cyclone of a 100 kW experimental Low Temperature Circulating Fluid Bed gasifier (Ahrenfeldt et al., 2013) at the Department of Chemical and Biochemical Engineering in the Technical University of Denmark. The original feedstock was dried sludge collected at the wastewater treatment plant in the Randers Municipality, Denmark, where P was captured approximately 50:50 by biological and chemical means. The chemical precipitation was based on Fe and Al chlorides.

2.2. Analytical methods and sampling

Small batches of around 200 g were sampled from the total mass collected. Each batch was dried at 105 °C overnight, cooled in the fume hood, homogenised with a steel spatula and immediately stored in plastic bags under dry conditions. A different batch was used for each experiment: alkaline extraction (Batch 1), two-stage chemical extraction (Batch 2) and ED experiments (Batch 3). The CaCO_3 content in another separate batch from the same overall sample was measured following the German standard DIN 18 129 (Rothenhofer et al., 2000).

The concentrations of elements were measured for each batch: in triplicates for the alkaline extraction as well as for the sequential extraction, and in eight samples for the ED experiments. It was also measured in triplicates after each step of the sequential extraction and the ED experiments. The targeted elements were: Al, As, Ca, Cd, Cu, Cr, Fe, K, Mg, Ni, P, Pb and Zn. Na was analysed only for the initial samples of the ED experiments. Their content was measured by Varian 720-ES inductively coupled plasma optical emission spectrometry (ICP-OES) after pre-treatment by Danish Standard DS259 (Danish standard, 2003): 1 g ash and 20 mL 7.3 M HNO_3 were heated at 200 kPa (120 °C) for 30 min. The liquid was thereafter separated by vacuum filtration through a 0.45 μm filter and diluted to 100 mL with deionised (DI) water.

Element concentrations among the batches were compared through a one-way analysis of variances (ANOVA).

2.3. Alkaline extraction

In order to assess the dependence of solubility of the target elements to alkaline solutions, 5 g of ash was shaken at 150 rpm for 1 week with 25 mL of NaOH at 1, 0.5, 0.1, 0.05 and 0.01 M respectively. The pH of the leachate was measured by a Radiometer Analytical pH electrode. The concentrations of the targeted elements in

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