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Electrodialytic treatment of municipal wastewater and sludge for the removal of heavy metals and recovery of phosphorus



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ABSTRACT

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Keywords: electrodialysis wastewater treatment sewage sludge heavy metals phosphorus Municipal wastewater and sewage sludge is an abundant source of phosphorus (P), but its usage is often limited due to wastewater treatment methods and contaminants, mostly heavy metals (HM's). Three compartment (3C) electrodialysis (ED) was used to simultaneously extract HM's (Cd, Cr, Cu, Ni, Pb and Zn) and recover P from municipal sludge samples obtained at different stages during wastewater treatment involving biological and chemical treatment as well as polymer addition for thickening of sludge and anaerobic digestion of excess sludge. Direct P recovery was investigated for high P reject water stream using the 3C ED cell setup and a two-compartment (2C) where the cathode in direct contact with the wastewater while P was extracted to and concentrated in the anolyte. Simultaneous extraction of HM's and recovery of P from wastewater or raw sludge using 3C ED was be most effective at a low pH using anaerobically digested sludge. The hydrolysis of OM during anaerobic digestion and the anaerobic conditions allowed for easier extraction of HM's such as Cd, Ni and Zn as they had fewer adsorption places, and improved P availability and extractability. Extraction of P from high concentration P streams was most effective using a 3C ED cell setup, with the electrodes separated from the sample by ionexchange membranes. Extraction with the 2C ED cell setup was less effective due to a rise in pH, caused by half reactions at the cathode and subsequent precipitation of P. For either removal of heavy metals or recovery of phosphorus using ED, the end-products in wastewater treatment, like anaerobically digested sludge and reject-water streams, are therefore best to be treated.

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1. INTRODUCTION

Phosphorus is an essential nutrient for the growth of organisms and cannot be substituted by any other element in nature. With current and projected consumption rates the primary source of phosphorus, mined phosphorus rock, is expected to run out within a range estimated between 50 to 400 years from now [1–3]. Meanwhile the quality of the mined phosphorus continues to decrease with an increased amount of impurities present [3]. To sustain the global demand for phosphorus, the need for and drive to obtain sustainable, secondary sources will significantly increase. For example, the Danish government aims to recover 80% of all phosphorus from municipal sludge by 2018, either through usage of sewage biosolids in agriculture, or by phosphorus recovery technologies [4]. The Swedish government adopted a policy with a national target of recovering and recycling 75% of phosphorus from

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http://dx.doi.org/10.1016/j.electacta.2015.04.097 0013-4686/© 2015 Elsevier Ltd. All rights reserved. sewage [5]. Municipal wastewater and sewage sludge is a high potential, secondary source of phosphorus and other nutrients. Optimal usage of phosphorus from these sources however, for example as fertilizer of agriculturals soil using sewage sludge, is still inhibited by the presence of organic and inorganic contaminants [6,7] as well as insoluble phosphorus complexes [8].

1.1. Heavy metal removal.

Even with strict regulations on chemical waste disposal in Denmark and many western countries, not to mention the general disregard of waste separation in developing countries, organic and inorganic contaminants can severely pollute sewage sludge [7]. This can negatively affect biological treatment [9] but also makes distribution or recycling of the available nutrients in the sewage sludge problematic. Instead of being used in agriculture, the polluted sewage sludge has to be disposed of in an alternative manner, for example through thermal treatment or disposal in landfill sites [6]. Although incineration can remove most organic contaminants [7,10] many of the inorganic contaminants, in the





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form of heavy metals, remain present in the resulting sewage sludge ash (SSA) [11].

Elektrokinetic remediation has been studied extensively for the remediation of heavy metal polluted media such as soils, sediments and (industrial) waste-streams [12], where remediation of municipal sewage sludge has mostly been studied for dewatered, anaerobically digested sludge [13–17]. Although these studies consider the effect of pH on HM removal, they do not explore the treatment of raw sludge as obtained directly from the wastewater treatment plant (WWTP). The studies generally do not consider changes in the sludge during wastewater treatment either, such as changes in organic matter (OM) content and pH, and additional biological processes which could potentially increase/reduce HM mobility and EK remediation efficiency.

1.2. Direct phosphorus recovery.

The formation of insoluble phosphorus complexes, the result of existing wastewater treatment methods, also limits the usage of phosphorus. Whenever biological processes do not sufficiently remove phosphorus from wastewater, additional chemical precipitation is applied [18,19]. Using iron and aluminum salts, aqueous phosphorus in the form of ortho-phosphate, is precipitated and removed with the sludge. Up to 95% of the existing wastewater treatment plants (WWTP's) in Denmark use some form of chemical precipitation to supplement biological phosphorus removal, resulting in up to 38% of the total phosphorus removed through chemical precipitation [20]. The formation of insoluble iron- and aluminum-phosphates is thought to decrease the crop-availability of phosphorus from sludge and sewage sludge ash (SSA) [8]. More so, after incineration of the sludge and obtaining an iron or aluminum rich SSA, strong acids are required to mobilize phosphorus [17,21]. When phosphorus is mobilized from SSA originating from sludge that was only subjected to enhanced biological phosphorus removal, with no additional chemical precipitation, boiling water was shown to be sufficient to mobilize almost all phosphorus [22].

To avoid usage of iron- or aluminum precipitates during wastewater treatment, we suggest removal of phosphorus during treatment using electrokinetics. Phosphorus is generally present in three different forms when entering a WWTP, as ortho-phosphate (ortho-P), poly-phosphate (poly-P) or as organic phosphates (org-P) (Fig. 1) [23,24]. Through biological and chemical treatment, phosphorus is bound to the sludge fraction leaving the WWTP

while the resulting effluent contains virtually no phosphorus and can safely be disposed in environmental waters.

Direct removal of phosphorus from wastewater through electrokinetics has successfully been achieved through electrocoagulation [25], a process that uses 'degradable' electrodes to chemically bind phosphorus. However, these phosphate precipitates are similar to those obtained after chemical precipitation using iron or aluminum salts, reducing crop-availability of phosphorus in the resulting sludge. Another form of electrokinetics able to remove phosphorus during wastewater treatment can be electrodialysis (ED).

ED can be used to directly recover and concentrate phosphorus in the form of ortho-phosphate (H_xPO4^{x-3}) , while minimizing interference with system processes or conditions in the sludge, as a significant change in pH, practical for mobilizing phosphorus, can also reduce biological activity. Previously application of ED in wastewater treatment has mostly been in the form of tertiary treatment for the removal of residual phosphorus and nitrogen from the effluent when concentrations exceeded legislative concentrations [19]. Although effective, low phosphorus concentrations made ED cost-intensive compared to other treatment methods [26-28]. ED treatment of undiluted sources, removing phosphorus and ammonium from black and grey water, has proven to be successful [29,30], but implementation of treatment methods at such an early stage and large scale would require extensive and costly adjustments to existing wastewater gathering systems [31]. The most economical feasible place for any technique aimed at recovering phosphorus during wastewater treatment, and leaving it in a crop-available form, would be to use the phosphorus-rich side streams or process waters with phosphorus concentrations >50 mg/L [32]. These conditions can mostly be found after anaerobic treatment processes, for example during nitrification/ denitrification processes and after anaerobic digestion. Besides the fact that the obtained concentrated phosphoric acid can be used in a wide range of applications, supplementing biological phosphorus removal through ED could reduce or effectively remove chemical precipitation completely.

The purpose of this work is to investigate the effectiveness of simultaneous extraction of heavy metals and removal of phosphorus using ED during wastewater treatment. Wastewater and sludge samples are taken at different stages during wastewater treatment, analyzed for different parameters and subjected to electrodialytic extraction experiments. Furthermore, for direct recovery of phosphorus aimed to reduce the usage of chemical precipitants,

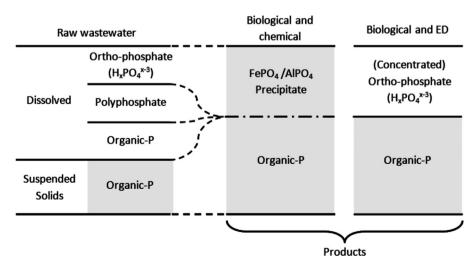


Fig. 1. Phosphorus speciation in liquid (dissolved) and solid (suspended solids) as it enters (raw wastewater) and, after a combination of biological/chemical or biological/ED treatment, exits (products) the wastewater treatment plant.

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