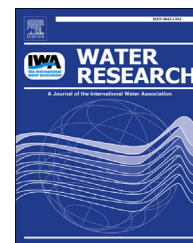


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Electrocoagulation treatment of peat bog drainage water containing humic substances

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ARTICLE INFO

Article history:

Received 10 March 2015

Received in revised form

13 April 2015

Accepted 17 April 2015

Available online 5 May 2015

Keywords:

Electrocoagulation (EC)

Techno-economic analysis

Large-scale experiments

Peat bog drainage water

Humic substances

ABSTRACT

Electrocoagulation (EC) treatment of 100 mg/L synthetic wastewater (SWW) containing humic acids was optimized (achieving 90% COD_{Mn} and 80% DOC removal efficiencies), after which real peat bog drainage waters (PBDWs) from three northern Finnish peat bogs were also treated. High pollutant removal efficiencies were achieved: P_{tot}, TS, and color could be removed completely, while N_{tot}, COD_{Mn}, and DOC/TOC removal efficiencies were in the range of 33–41%, 75–90%, and 62–75%, respectively. Al and Fe performed similarly as the anode material.

Large scale experiments (1 m³) using cold (T = 10–11 °C) PBDWs were also conducted successfully, with optimal treatment times of 60–120 min (applying current densities of 60–75 A/m²). Residual values of Al and Fe (complete removal) were lower than their initial values in the EC-treated PBDWs.

Electricity consumption and operational costs in optimum conditions were found to be low and similar for all the waters studied: 0.94 kWh/m³ and 0.15 €/m³ for SWW and 0.35–0.70 kWh/m³ and 0.06–0.12 €/m³ for the PBDWs (large-scale). Thus, e.g. solar cells could be considered as a power source for this EC application. In conclusion, EC treatment of PBDW containing humic substances was shown to be feasible.

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1. Introduction

Peat is one of the most important energy sources in Finland; annually, peat covers about 7% of the energy supply and 20% of district heat (Geological Survey of Finland, 2015). Globally, it is estimated that 3% of the total landmass is peatland, and the

main producers and users of peat are Finland, Belarus, Estonia, Ireland, Indonesia, Sweden, and the Russian Federation (World Energy Council, 2014). Most of these countries lie in the Northern Hemisphere, thus they have a long and cold winter season. There is an ongoing debate on whether peat energy should be produced or not, considering, e.g. its effect

Abbreviations: COD_{Mn}, chemical oxygen demand [mg/L]; DC, direct current; DOC, dissolved organic carbon [mg/L]; EC, electrocoagulation; EEC, electrical energy consumption [kWh/m³]; EMC, electrode material consumption [kg/m³]; HA, humic acids; HS, humic substances; i, current density [A/m²]; ICP, inductively coupled plasma; OC, operating costs [€/m³]; OES, optical emissions spectrometer; PBDW, peat bog drainage water; SEC, supporting electrolyte consumption [kg/m³]; SWW, synthetic wastewater; TOC, total organic carbon [mg/L]; TS, total solids [mg/L].

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<http://dx.doi.org/10.1016/j.watres.2015.04.029>

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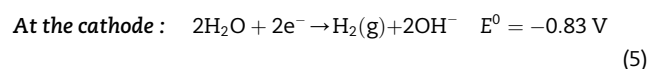
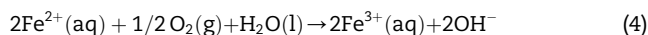
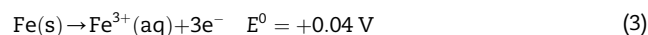
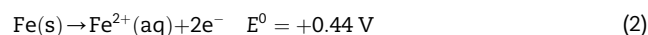
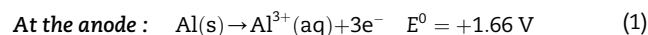
on global warming and on the other hand its domesticity and vast reserves. However, one of the main problems of peat energy production (especially in Finland) is peat bog drainage water (PBDW), which can even be considered one of the key questions affecting the future of the entire Finnish peat industry due to the increasingly strict environmental regulations being proposed. To meet these regulations, new ideas are needed, since the effectiveness of conventional water treatment methods is often limited.

PBDW is typically slightly acidic and colored, and it contains nutrients (P and N) as well as humic substances (HS) and total solids (TS). Excessive levels of nutrients are considered detrimental in natural bodies of water because oversaturation causes algae growth, leading to eutrophication (Bektaş et al., 2004; Zhao and Sengupta, 1998; Alvarez-Vázquez et al., 2014). This in turn depletes the oxygen level and hinders penetration of light into the water, thus adversely affecting organisms present in the aquatic environment and reducing biodiversity (Bektaş et al., 2004; Alvarez-Vázquez et al., 2014). HS occur in soil and natural waters as residues of plant and animal decay by microbial activity—most organic matter found in natural waters is constituted of HS (Ghernaout et al., 2009; Jones and Bryan, 1998; Seida and Nakano, 2000; Yildiz et al., 2007).

One of the main components of HS in water (along with fulvic acids and humins) are humic acids (HA), which are weakly acidic aliphatic and aromatic compounds containing functional groups such as $-\text{COOH}$ and phenolic $-\text{OH}$ groups (Prado and Airolidi, 2003; Seredyńska-Sobecka et al., 2006; Naddeo et al., 2007; O'Melia et al., 1999). These acidic compounds add a dark color to natural waters (Motheo and Pinheiro, 2000). This causes esthetic problems and further hinders the availability of light in the water. HS are heterogeneous in structure, and on the whole their chemical structures are very complex with no defined physicochemical properties and a high molecular weight (several hundreds or larger) (Motheo and Pinheiro, 2000; Hesse et al., 1999). They are highly stable, and therefore are retractive to attack by microorganisms (Motheo and Pinheiro, 2000). Although they are at the end-point of nature's biodegradative and oxidative process (virtually non-biodegradable), due to their high aromatic and aliphatic residue content HS can readily be aggregated and precipitated by charge neutralization (Jones and Bryan, 1998; Yildiz et al., 2007; Ødegaard et al., 1999; Yildiz et al., 2008). Other potential methods for HS removal may include physicochemical processes, biological processes, membrane processes, etc., and especially classical methods such as usage of peatland buffer areas and/or wetlands.

Electrocoagulation (EC) is a water treatment technology that has been known for over a hundred years now, but it is currently under intensive development and of commercial interest (Chen, 2004; Kuokkanen et al., 2013). In EC, so-called sacrificial anodes (commonly Al or Fe) are dissolved into water *in situ*, promoting coagulation, while microscopic hydrogen gas is usually generated simultaneously at the cathode, promoting flotation. The only chemical species used in EC is the electron (excluding possible pH and conductivity alteration chemicals), thus making EC a green technology. Charge neutralization of negatively charged colloids by cationic hydrolysis products and sweep flocculation

(enmeshment of pollutants in the amorphous hydroxide precipitate produced) are proposed as the main functional mechanisms of Al and Fe in EC (Karhu et al., 2012). The principal electrochemical reactions in the EC process are presented in Eqs. (1)–(5):



In principle, as presented in Eq. (3), iron may oxidize directly to Fe^{3+} at the anode, but this reaction is highly unfavorable ($E_0 = +0.04 \text{ V}$) compared with oxidation to ferrous (Fe^{2+}) iron ($E_0 = +0.44 \text{ V}$), as presented in Eq. (2). Dissolved oxygen in solution (see the sum reaction presented in Eq. (4)) may cause oxidation of the electrogenerated Fe^{2+} to Fe^{3+} (Zodi et al., 2009). The electro-generated Al^{3+} or Fe^{3+} ions undergo immediate spontaneous reactions in which corresponding hydroxides and/or polyhydroxides are produced (Mollah et al., 2004). The mass of the dissolved metal [g] can be calculated theoretically from Faraday's law (Eq. (6)), although the theoretical amount of anodic dissolution is found to be exceeded in many real EC applications (superfaradaic efficiencies).

$$m_{\text{metal}} = \frac{ItM}{zF} \quad (6)$$

where I is the applied current [A], t is the treatment time of the EC process [s], M is the molar mass of the anode metal [g/mol], z is the valence number of ions of the substance ($z_{\text{Al}} = 3$, $z_{\text{Fe}} = 2$), and F is Faraday's constant (96,485 C/mol).

The experimental values of superfaradaic anode metal dissolution have varied case-specifically between 105% and 190% of the theoretically expected value (Terrazas et al., 2010; Kongjao et al., 2008; Şengil and Özacar, 2009; Kobya et al., 2011; Yetilmezsoy et al., 2009; Mouedhen et al., 2008; Kuokkanen et al., 2015). It has been proposed that this phenomenon is due to pitting corrosion, especially in the presence of chlorine ions (Chen, 2004). In addition, when chloride is present in solution (e.g. from NaCl) and the anode potential is sufficiently high, active chlorine species (Cl_2 , HClO , OCl^-) may form. These species may then both oxidize pollutants and simultaneously chemically oxidize the anode material, and thus enhance the performance of an EC reactor. Moreover, Fe^{2+} can reduce organics to be transformed into Fe^{3+} . Superfaradaic anode dissolution is a parameter of whose effect on the EC process is rather rarely studied and should be taken into account when analyzing the economics of a given EC application, since metal material and electricity costs are regarded as the main cost components of EC.

Even though the use of EC in treating various types of water and wastewater has recently been studied extensively, to the best of our knowledge there are no previous scientific papers dealing with EC treatment of PBDW. Also, scale-up studies on

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