



# Electrochemical oxidation of iron and alkalinity generation for efficient sulfide control in sewers



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## ABSTRACT

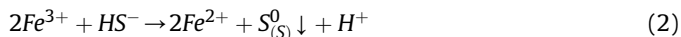
The addition of iron salts is one of the most commonly used dosing strategies for sulfide control in sewers. However, iron salts decrease the sewage pH which not only reduces the effectiveness of sulfide precipitation but also enhances the release of residual sulfide to the sewer atmosphere. Equally important, concentrated iron salt solutions are corrosive and their frequent transport, handling, and on-site storage often come with Occupational Health and Safety (OH&S) concerns. Here, we experimentally demonstrated a novel sulfide control approach using electrochemical systems with parallel placed iron electrodes. This enabled combining anodic dissolved iron species release with cathodic hydroxyl anion production, which alleviates all the aforementioned concerns. A long-term experiment was successfully carried out achieving an average sulfide removal efficiency of  $95.4 \pm 4.4\%$  at low voltage input of  $2.90 \pm 0.54$  V over the course of 8 weeks. This electrochemical method was demonstrated to successfully achieve efficient sulfide control. In addition, it increases the sewage pH, thereby overcoming the drawbacks associated with the pH decrease in the case of conventional iron salt dosing. Ferrous ions were produced at an overall coulombic efficiency (CE) of  $98.2 \pm 1.2\%$ , whereas oxygen evolution and direct sulfide oxidation were not observed. Short-term experiments showed that increasing either inter-electrode gap or current density increased the cell voltage associated with the increase in the ohmic drop of the system. Overall, this study highlights the practical potential of in-situ generation of dissolved iron species and simultaneous hydroxyl anion generation for efficient sulfide control in sewers.

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## 1. Introduction

Hydrogen sulfide generation by sulfate reducing bacteria (SRB) in sewer pipes is a notorious problem in sewer management globally (Hvitved-Jacobsen et al., 2013; Pikaar et al., 2014). Conventional sulfide abatement strategies mainly involve the addition of chemicals to either prevent the formation of hydrogen sulfide or mitigate the effect of hydrogen sulfide generated (Zhang et al., 2008). The addition of iron salts, nitrate, sodium hydroxide, magnesium hydroxide and oxygen are the five most commonly used chemicals for sulfide control by water utilities (Ganigue et al., 2011). The working principle of iron salt dosing is that ferrous ions ( $\text{Fe}^{2+}$ )

precipitate sulfide to form ferrous sulfide ( $\text{FeS}$ ) according to equation (1), while ferric ions ( $\text{Fe}^{3+}$ ) chemically oxidize sulfide to elemental sulfur ( $\text{S}^0$ ) according to equation (2). The reduced  $\text{Fe}^{2+}$  can subsequently precipitate sulfide (Firer et al., 2008; Nielsen et al., 2005).



Iron salt dosing is suitable for any size of sewer pipes as sulfide precipitation occurs in the bulk liquid (Ganigue et al., 2011) and it is effective in controlling dissolved sulfide concentration in sewers to very low levels (i.e.  $< 1$  mg S/L) (Nielsen et al., 2005). Although a proven strategy that has been used by water utilities globally for decades, it comes with some disadvantages (WERF, 2007). Ferric

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salts are strongly acidic compounds (e.g.  $pK_a = \text{Fe}(\text{H}_2\text{O})_6^{3+}$  is 2.20) (Brezonik and Arnold, 2011; Cornell and Schwertmann, 1996) and thus conventional ferric dosing results in a pH decrease in sewage (WERF, 2007; Zhang et al., 2016). Sulfide precipitation reaction is highly pH dependent. It is negatively affected by lower pH, especially around the most prevalent circumneutral sewage pH levels (i.e. 6–8) (Firer et al., 2008; Hvitved-Jacobsen et al., 2013; Nielsen et al., 2008). For example, a decrease from pH 7.0 to 6.5 resulted in an increase in iron dosing requirements of more than 200% in order to achieve the same sulfide concentrations of 0.1 mg S/L (Firer et al., 2008). In addition, a decrease in sewage pH also shifts the equilibrium from  $\text{HS}^-$  to the volatile  $\text{H}_2\text{S}$ , releasing more residual hydrogen sulfide to the sewer atmosphere in gravity sections which could accelerate sewer corrosion in sections downstream of the dosing point (WERF, 2007). Moreover, the frequent transport, handling, and storage of purchased iron salt solutions are associated with Occupational Health and Safety (OH&S) concerns as it is a highly corrosive and hazardous compound (WERF, 2007). Therefore, the delivery and storage systems are usually equipped with special materials and equipment to meet their safety standards (WERF, 2007).

In-situ generation of dissolved iron species and hydroxyl anions in sewers could potentially overcome OH&S concerns associated with purchased iron salts. In addition, cathodically generated hydroxyl anions could increase the sewage pH, thereby avoiding the acidifying effect of iron salt dosing. Although it is theoretically feasible to generate dissolved iron species and hydroxyl anions in sewage on-site, coagulation could potentially occur while electrochemically dissolving iron in sewage which hinders its practical sewer application. Coagulation is the core mechanism of electrocoagulation (EC), a widely used electrochemical process to remove pollutants from water and wastewater (Canizares et al., 2008; Kobya et al., 2003; Vasudevan et al., 2008; Zodi et al., 2013). This results in solids agglomeration and sludge generation (Mollah et al., 2001; Sahu et al., 2014; Vong and Garey, 2014). For example, electrocoagulation has been demonstrated to efficiently remove hydrogen sulfide from tannery wastewater, however, also resulted in efficient chemical oxygen demand (COD) removal (i.e. > 90% removal) and sludge generation associated with the high COD removal (Muruganathan et al., 2004). The extra sludge produced could enhance sewer sediment formation in the slower flowing gravity sections downstream of the dosing location (George et al., 2003).

In the EC process, the removal of pollutants becomes faster as well as more efficient at a higher current density due to the increase in both the metal dissolution rate (i.e. coagulant dosage rate) and hydrogen bubble production rate (Adhoum et al., 2004). The increase in metal dissolution rate generates more coagulants (Sahu et al., 2014) while the increased gas production rate creates a stronger upward flux (i.e. better solution mixing), leading to increased pollutant removal by flotation (Holt et al., 2002). In addition to current density, physical mixing of the treated solution affects the coagulation process. The coagulants generated and pollutants are brought together to form solid agglomerates by physical mixing and as such rapid physical mixing is normally employed to enhance coagulation (Vong and Garey, 2014). Therefore, we hypothesize that by applying a low current density and minimizing physical mixing, solids aggregation can be avoided and thus formation of solids occurs after the cell operation, and not in it.

In addition to coagulation, in an electrochemical system containing water and sulfide, reactions like direct oxidation of water or sulfide could occur if the anode potential is sufficient, leading to the formation of oxygen and elemental sulfur, respectively (Dutta et al., 2010; Muruganathan et al., 2004; Pikaar et al., 2011, 2012; Townsend, 2014). While a sacrificial iron anode is dissolved in

sewage for sulfide control, these secondary water and/or sulfide oxidation reactions compete with iron dissolution, resulting in increased electricity consumption to achieve the desired iron dosage. Additionally, the generation of oxygen further contributes to solution mixing and thus promotes coagulation. Equally important, direct sulfide oxidation on the anode could further lead to anode passivation by elemental sulfur deposition, decreasing the longevity of the electrochemical process (Dutta et al., 2008).

In this study, we applied the basic principle of EC to produce dissolved iron species (i.e.  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ ) and hydroxyl anions in sewage but operated the electrochemical cell with essential differences. We applied a low current density and slow physical mixing to avoid/minimize coagulation, one of the core mechanisms of EC process, thereby minimizing agglomeration of solids while dissolving iron. The dissolved iron species generated are expected to precipitate or react with sulfide present in sewage while the produced hydroxyl anions are expected to increase the sewage pH. Compared to EC, solid-liquid separation is not required which alleviates operational constraints.

In this proof of concept study, we aim to demonstrate in-situ electrochemical oxidation of iron coupled with hydroxyl anions generation in sewage for sulfide control, thereby overcoming the drawbacks of conventional iron salt dosing. In this study, the dissolved iron species generated (i.e.  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ ) and the overall Coulombic efficiency (CE) for iron dissolution were determined. Continuous experiments over a period of 8 weeks were conducted to demonstrate its long-term sulfide removal effectiveness and stability of the process in terms of energy requirements. Subsequently, short-term experiments were conducted using different current-to-flow ratios to achieve different electron/S ratios to investigate the impact of electron/S dosing ratios on sulfide removal efficiency and sewage characteristics in terms of pH as well as the removal of COD from sewage. Finally, the impacts of inter-electrode gap and applied current density on the energy requirement of the system were investigated.

## 2. Material and methods

### 2.1. Electrochemical cell and experimental setup

A schematic overview of the experimental setup is given in Fig. 1. Experiments were conducted using single-compartment electrochemical cells made of 600 mL round plastic container, 100 or 200 mL polypropylene measuring cylinders, depending on the experiment conducted. Fresh sewage was collected from a local pumping station in Brisbane on a weekly basis and immediately stored at 4 °C. A water bath was used to heat up the sewage to  $21 \pm 1$  °C and a concentrated sulfide stock solution (prepared using  $\text{Na}_2\text{S} \cdot 9\text{H}_2\text{O}$ , Sigma-Aldrich) was added to the sewage using a syringe pump (NE-1000, New Era Pump Systems, Inc., USA) before sewage being fed to the electrochemical cell at influent sulfide concentrations of 10 or 15 mg S/L, depending on the experiment conducted. A peristaltic pump (Masterflex pump) was used to control the sewage flow rate at 1 L/h for all experiments. Iron plates (mild steel flat bar, Total Steel Australia) were used as both anode and cathode. The dimensions of iron plates were 12.8 cm  $\times$  3.8 cm  $\times$  0.9 cm in the 600 mL reactor and 20 cm  $\times$  1.3 cm  $\times$  0.3 cm in 100 and 200 mL reactors. Anode and cathode were facing each other with an inter-electrode gap distance of 1.2 cm. Electrodes were submerged in sewage at different depths to create different geometric electrode surface areas. Both electrodes were galvanostatically controlled using a Wenking potentiostat/galvanostat (KP07, Bank Elektronik, Germany). A fixed current density of 2 mA/cm<sup>2</sup> was applied in all experiments unless stated otherwise. A data acquisition unit (Agilent Technologies, USA) or constant-current chronopotentiometries

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