



# Electrochemical decrease of sulfide in sewage by pulsed power supply



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## ABSTRACT

Pulsed electrochemical process was explored to remove sulfide in municipal sewage, which successfully achieved higher removal rate of 93.2% compared with 73.2% by direct current (DC) power supply. The sulfur content on the electrode was 0.21% by pulsed electrochemical process compared with that of 1.05% on anode by DC electrochemical process, indicating that pulsed electrochemical process can inhibit anodic passivation resulting from sulfur deposition compared with DC electrochemical process. Besides, the electrode scaling can also be effectively controlled during pulsed electrochemical process. Moreover, the process optimization was investigated for pulsed electrochemical treatment, which obtained the highest removal rate of 97.8% at pH 7–8, cell voltage of 7 V, duty cycle of 60%, pulse frequency at 1000 Hz and electrode spacing of 2.5–3.0 cm. These results indicate that pulsed electrochemical process is a promising method of efficient and sustainable electrochemical sulfide removal from sewage.

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## 1. Introduction

Municipal underground sewer networks are fundamental infrastructures in cities. The erosion caused by internal or external corrosive medium, may seriously damage the underground concrete sewage pipes, resulting in ground subsidence, sewage leakage or other accidents. Sulfide emission is associated with biogenic corrosion of sewage pipes, emergent obnoxious odors in urban atmosphere and toxicity of sulfide gas to sewer workers [1]. Generally, sulfide should be eliminated from the wastewater or exhaust gas before discharge. Since a dominant source of sulfide in sewage pipes is the reduction of sulfate by sulfate reducing bacteria (SRB) located in the biofilms of anaerobic sewer sections, it is widely required to control the sulfide in sewers for preventing municipal pipelines corrosion.

Electrochemical techniques are alternative methods for wastewater treatment due to its versatility, environmental compatibility and potential cost-effectiveness [2]. Removal of sulfide from wastewater employing electrochemical processes has been demonstrated in previous studies [3–5], which sulfide can be directly oxidized at the anode [6,7]. Elemental sulfur has been found the dominant oxidation product when carbon/graphite materials were used as the electrode [8]. However, the main

drawbacks of electrochemical sulfide removal are anodic passivation by the precipitated elemental sulfur [9,10] and cathodic polarization as a result of scale deposition [11].

Sulfide extraction with organic solvent followed by solvent evaporation, and controlled sulfide precipitation in the bulk solution with saturated polysulfides under alkaline conditions at elevated temperatures have been proven to delay the electrode passivation or regenerate electrodes [12,13]. However, both methods are not considered sustainable due to high energy consumption and toxic organics presence. Dutta et al. proposed an *in situ* regeneration approach that continuously remove dissolved sulfide from wastewater at an anode as sulfur, meanwhile, the recovery of deposited sulfur was achieved via regular switching between anodic sulfide oxidation and cathodic sulfur reduction [14]. Besides, Panizza et al. applied multiple current steps electrolysis and a semi-continuous current control mode in the boron-doped diamond electrolysis that achieved the mineralization of organic pollutants with high reaction kinetics and current efficiency as well as low energy consumption [15].

Cathodic scaling or mineralization can hinder the electrochemical process [11]. Several methods were developed to control scale deposition on cathode in electrochemical cells including: adding anti-scalant [11], which may lead to secondary contamination and chemical consumption; the fluidized bed cathode [16], which is material and power consumptive; changing electrochemical processes [17,18], which increase difficulty in management. Alternating current (AC) [19–21] and alternating pulsed current

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(APC) [22–24] have been applied in electrocoagulation process, which is considered to retard the electrode scaling experienced with direct current (DC). Kamaraj et al. proved that AC had the same level of effect on copper removal from water by electrocoagulation process as DC with lower energy consumption, due to the prevention of oxide layer and corrosion on electrodes [25].

The pulsed current supply was demonstrated a feasible approach for organic removal by electrochemical oxidation, for the obvious specific energy consumption reduction was achieved compared with constant current supply [26]. Also, lower deposition rate of particles under pulsed current has been proved compared with DC [27]. Periodic reversal of the electrodes has been demonstrated effectively inhibits anodic passivation and cathodic polarization in electrosynthesis [28], electrocoagulation [29,30] and electrochemical oxidation [14]. Based on the aforementioned researches, in this study, pulsed current supply was employed, aiming to electrochemically remove sulfide from wastewater as well as *in situ* avoid anodic passivation derived from sulfur deposition and cathodic scaling. The influences of technical parameters (pH, cell voltage, pulse duty cycle, pulse frequency and electrode spacing) on the system performance were investigated and optimized as well. By means of energy dispersive X-ray spectrometry (EDS) and sulfide removal rates analysis, the superiority of pulsed current supply was evaluated compared with constant current supply. The morphology of deposits on electrodes was characterized with scanning electron microscopy (SEM).

## 2. Materials and methods

### 2.1. Simulated sewage

The sewage used in this research was collected from the sewers in the campus of ECUST, while  $150 \text{ mg L}^{-1}$  D-glucose,  $20 \text{ mg L}^{-1}$   $\text{NH}_4\text{Cl}$  and  $10 \text{ mg L}^{-1}$   $\text{Na}_2\text{S}$  were added to simulate the municipal sewage. The water quality of simulated sewage is shown in Table 1.

### 2.2. Electrochemical reactor and operation

A single-chamber electrochemical reactor (bottom size  $10 \times 10 \text{ cm}$ ) containing 1 L of simulated sewage was employed, which was supplied with the DC power (Jinbaili GKW-F, Daheng Electric, Jiangyin, China) or the pulsed power (Jinbaili DXN-F, Daheng Electric, Jiangyin, China). Two graphite plates ( $12 \text{ cm} \times 5 \text{ cm} \times 4 \text{ mm}$ ) were used as electrodes and submerged 9 cm in the bulk sewage. Using a stirring bar, the electrochemical reactor was located on a magnetic stirrer at a proper rotating speed and operated in fed batch mode (shown in Fig. 1). To determine the sulfide concentration, the sewage was sampled every 30 min during a total reaction time of 150 min. The experiment was replicated three times for accuracy. The mean and standard deviation of the data were calculated.

### 2.3. Analysis and spectrometry

Sulfide ( $\text{S}^{2-}$ ,  $\text{HS}^-$  and  $\text{H}_2\text{S}$ ) concentrations were quantified with the iodometric method, according to the national standard method of China for determination of sulfide content in water (HJ/T

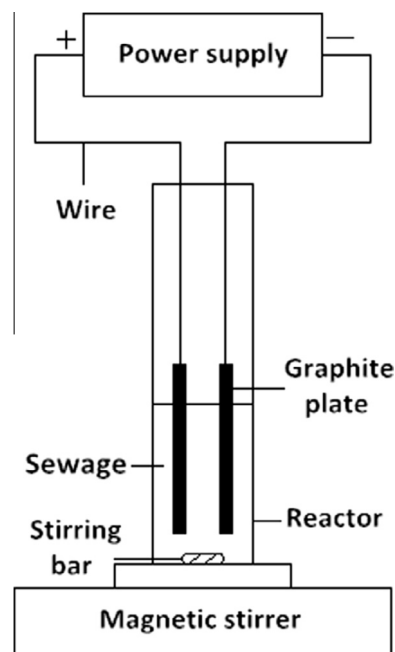


Fig. 1. Schematic of electrochemical reactor for sulfide removal.

60–2000). SEM images and EDS spectra of the graphite plates were performed to analyze the deposits on electrode surface using the scanning electron microscope (S-3400N, Hitachi, Tokyo, Japan) equipped with the energy-dispersive X-ray spectrometer (Genesis XM2, Ametek, San Diego, CA).

### 2.4. Experimental procedures

#### 2.4.1. Performance of pulsed power compared with DC power

The performance of electrochemical reactor was evaluated based on sulfur removal rate and sulfur deposition on the surface of electrode. Two electrochemical reactors of the same size were employed to remove sulfide from the sewage ( $\text{pH } 7.0 \pm 0.1$ ) with an electrode spacing of 2.5 cm. The reactors were operated at cell voltage of 3 V, supplied with the DC power and the pulsed power, respectively. The duty cycle and pulse frequency for the pulsed power were set as 50% and 1000 Hz. After the electrochemical process, the graphite plates were taken out to analyze the sulfur deposit and scale on the surface of electrodes through SEM and EDS.

#### 2.4.2. Optimization of operational parameters with pulsed power supply

The operational parameters that have influence on the electrochemical sulfide removal with pulsed power supply, *i.e.*, pH, cell voltage ( $U$ ), pulse duty cycle ( $D$ ), pulse frequency ( $f$ ) and electrode spacing ( $d$ ), were investigated and optimized.

## 3. Results and discussion

### 3.1. Performance of pulsed power compared with DC power

#### 3.1.1. Electrochemical sulfide removal in sewage

Both the concentrations of sulfide under DC power and pulsed power supply dropped from initial  $12.7$  to  $8.5 \text{ mg L}^{-1}$  in 30 min and appeared to be nearly identical. However, after half an hour, the rate of sulfur removal apparently had more increase with the pulsed power than that with the DC power over time. After the total reaction time (150 min), sulfide removal rate of the

Table 1  
Water quality of simulated sewage ( $\text{mg L}^{-1}$ ).

Chemical oxygen demand	Sulfide-S	Sulfate-S	Ammonium-N	Total N	Total P
$350 \pm 36$	$12.7 \pm 0.6$	$34.5 \pm 3.2$	$35.6 \pm 4.1$	$42.3 \pm 0.3$	$3.6 \pm 0.2$

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