



Communication

Advantage of selective production of green rusts for Sb(V) removal in Fe(0) electrocoagulation



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ABSTRACT

The removal efficiency of pollutants in Fe(0) electrocoagulation (EC) has been associated closely with the speciation of generated Fe(II)/Fe(III) oxides during this process, which is very complicated and can be affected by various factors. In this work, *in-situ* Raman, X-ray diffraction and some other techniques have been used to study the speciation of Fe under different conditions and to establish a relationship between Fe speciation and Sb(V) removal efficiency. Results indicated that concentration of dissolved oxygen (DO) is a key factor influencing Fe(0) EC. It was found that green rusts (GRs) were formed and were then transformed into magnetite at lower DO concentration, and Sb(V) removal efficiency reached 99.9% after 30 min of EC. In contrast, γ -FeOOH was formed at high DO concentration, and the removal efficiency of Sb(V) after 30 min of EC was only 72.8%. In the presence of sulfite and phosphate with low concentrations, GRs can be stabilized and benefit the removal of Sb(V). We believe this work will provide some new insights on the mechanism of Fe(0) EC and the effective removal of other pollutants during Fe(0) EC process.

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Antimony (Sb) is a widely used metal in industry. But it is also an element of increasing environmental concern due to its toxicity and potential carcinogenicity. In natural waters, Sb(III) and Sb(V) are the two main oxidation states of Sb, and Sb(V) is the predominant form in oxidized waters [1]. Techniques such as coagulation [2], ion exchange [3], membrane filtration [4,5], sorption [6–8], cathodic deposition [9], *etc.*, have been used for antimony removal, and it is generally believed that the removal of Sb(V) is more difficult than that of Sb(III) because of the high solubility of Sb(V) [10]. The study of Sb removal through chemical coagulation showed that coagulation was effective for Sb(III) removal, but its efficiency for Sb(V) removal was limited. Similar results have been obtained by Wu *et al.* [11]. Therefore, there is an urgent need for the development of an effective method for Sb(V) removal.

Electrocoagulation (EC) is attracting much recent attention because of its advantages such as convenience and low cost. It is accomplished through the *in-situ* generation of coagulants by electrolytic oxidation of the anode material and has been widely applied in the treatment of pollutants such as oil, dyes, heavy metals, bacteria, *etc.* [12–16]. A few reports have studied the removal of Sb by EC [17,18], which indicated Fe(0) EC was more effective than Al(0) EC for Sb removal. But these studies were not very detailed, and there were still many aspects remaining to be elucidated.

It has been reported that the Fe speciation is an important factor influencing the removal of pollutants in EC, and it can be affected by operating parameters such as current density, pH, dissolved oxygen (DO) and anode potential [19–21]. The reported species generated during Fe(0) EC include magnetite (Fe₃O₄), hematite (α -Fe₂O₃), ferrihydrite (5Fe₂O₃·9H₂O), maghemite (γ -Fe₂O₃), goethite (α -FeOOH), akaganeite (β -FeOOH), lepidocrocite (γ -FeOOH), green rusts (GRs, [Fe^{II}_(6-x)Fe^{III}_x(OH)₁₂]^{x+}[(A)_{x/n}·yH₂O]^{x-}), *etc.* [20–22]. However, there is a lack of a comprehensive understanding of the relationship between reaction condition and Fe speciation, and to the best of our

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knowledge, the impact of Fe speciation on the removal of Sb(V) has not been reported yet.

In this work, Sb(V) removal efficiency during EC was evaluated under different reaction conditions, especially under different pH values and concentrations of dissolved oxygen (DO). The main aims of this work include: (1) To establish a relationship between Fe speciation and Sb(V) removal efficiency; (2) to identify the key species that are more efficient for Sb(V) removal and the selective generation of these species in Fe(0) EC; (3) to investigate into the possible mechanism of Sb(V) removal in Fe(0) EC. Different operation parameters were also optimized to obtain a higher Sb(V) removal efficiency.

EC experiments were conducted under high and low DO concentration conditions. For low DO concentration experiment, the cell was purged with N₂ until the concentration of DO dropped to below 0.3 mg/L, and the experiments were conducted under N₂ atmosphere. For high DO concentration experiments, the cell was purged with O₂ for 30 min before each experiment and was kept under constant purging of O₂. Initial solution pH was adjusted with 0.1 mol/L H₂SO₄ and 0.1 mol/L NaOH. The electrolyte was 5 mmol/L Na₂SO₄ with a Sb(V) concentration of 0.5 mmol/L (about 60 mg/L). All solutions were prepared with Milli Q water (18.2 MΩ cm). The electrodes were connected to a DC power supply in galvanostatic mode. Samples were collected and kept in 4 °C before analysis. The theoretical value of the mass of generated Fe in EC could be calculated by Faraday's law of electrolysis according to the current density [19,20]. Total Sb and Fe concentrations were determined using an inductively coupled plasma optical emission spectrometer (ICP-OES) (SCIEX Perkin Elmer Elan mode 5000) after dissolving the sample with 0.1 mol/L HCl. Sb and Fe concentration in aqueous phase were measured by filtering the sample through a 0.22 μm filter. DO and pH variations were measured by a DZB-718 portable multi-parameter meter (Shanghai REX Instrument Factory, China). X-ray diffraction (XRD) measurements were performed on powder samples using Cu Kα radiation on a Panalytical X'Pert Pro diffractometer. The samples sensitive to oxidation were mixed with a drop of glycerol after filtration before XRD analysis. Transmission electron microscopy (TEM) images were taken by JEM 2100 F (JOEL, Japan) equipped with energy dispersive X-ray analysis (EDAX). Raman spectra were taken with LabRAM HR800 laser confocal Raman spectrometer equipped with a wet objective (HORIBA Jobin Yvon, France).

The process of Fe(II) oxidation in EC is one of the most important factors influencing the formation and the transformation of flocs and the removal efficiency of pollutants during EC, which is strongly related to the concentration of DO and the solution pH [23,24]. In this work, first of all, the dependence of Sb(V) removal under high and low DO concentrations at different initial pH values was studied at the pH range of 5.0–11.0, and results are presented in Fig. 1. As shown in Fig. 1A, Sb(V) removal efficiency reached 96.1% after 30 min treatment at initial pH of 5.0

under low DO concentration. Sb(V) removal efficiency after 30 min of EC at the initial pH of 5.0, 7.0 and 9.0 was nearly identical. At the initial pH value of 11.0, Sb(V) removal efficiency dropped to below 90% at 30 min. But it was still higher than the removal efficiency of Sb(V) under high DO concentrations. The decrease of Sb(V) removal efficiency was probably caused by the passivation of the anode, since at elevated pH, a passivation layer would form on the Fe anode, and the dissolution of Fe ions were inhibited, thus leading to a lower amount of the generated flocs in EC. The removal of Sb(V) under high DO concentration is shown in Fig. 1B. Sb(V) removal efficiency remained almost unchanged within the initial pH range of 5.0–9.0, and decreased to 65.7% at the initial pH of 11.0. It can be seen that the removal of Sb(V) under high DO concentration was lower than that at low DO concentrations, further proved that lower concentration of DO was more beneficial for Sb(V) removal in Fe(0) EC. To understand this difference, the variation of DO concentration and the generation of Fe under different DO conditions at the initial pH of 5.0 was also measured (Fig. S1A in supporting information). For high DO conditions, the concentration of DO remained relatively high (> 4.0 mg/L) after 30 min of EC, indicating the generated Fe(II) was completely oxidized to Fe(III), and the main form of the precipitates were Fe(III) (hydro)oxides. The slight decrease of DO was caused by the consumption of generated Fe(II) and the oxygen reduction reaction on the cathode [20]. Under low DO concentration condition, without the constant O₂ supply, DO dropped to 0 mg/L during EC within 5 min, which can be ascribed to the oxidation of generated Fe(II) and oxygen reduction on the cathode. Under this circumstance, the oxidation of Fe(II) to Fe(III) was significantly inhibited. The amounts of generated Fe were nearly equal at both high and low DO concentrations (Fig. S1B in supporting information), which indicated that the total amount of the formed flocs was not the main reason causing the difference in Sb(V) removal efficiency.

The above results have shown that low DO concentration is more favorable for the removal of Sb(V) in EC. Meanwhile, other factors such as current density and the concentration of Sb(V) can also affect the performance of EC. So we further optimized these operation parameters to achieve a higher Sb(V) removal efficiency. Results showed increased removal efficiency with increasing current density (Fig. S2A in Supporting information). From Faraday's laws of electrolysis, it can be seen that the amount of generated Fe is in proportion to the current density in the EC process, and higher current density can lead to faster generation of flocs, thus facilitating the removal of Sb(V) during EC process. The influence of initial Sb(V) concentration is presented in Fig. S2B (Supporting information), which indicated that the removal efficiency of Sb(V) decreased with increasing Sb(V) concentration. When the initial concentration of Sb(V) was below 50 mg/L, Sb(V) could be effectively removed within 30 min of retention time. However, when Sb(V) concentration was above 100 mg/L, a significant decrease in Sb(V) removal efficiency is observed, which requires extended treatment time.

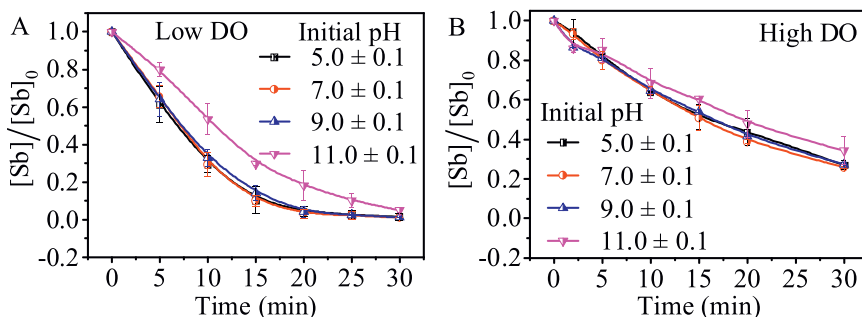


Fig. 1. The efficiency of Sb(V) removal as a function of pH under different DO conditions. (A) Low DO condition. (B) High DO condition.

As one of the most important influencing factors for Fe(0) EC, the effect of DO concentration on the flocs generated in Fe(0) EC were investigated with *in-situ* Raman, XRD and TEM analysis. Raman analysis is a quite useful tool for the characterization of Fe (hydro)oxides [21,22]. The flocs formed under different conditions at different time intervals were further characterized by Raman analysis and results are presented in Figs. 2A and B. For the flocs formed at low DO concentration, at 10 min, three peaks at 423, 507 and 675 cm^{-1} were observed (Fig. 2A), the Raman shift at 423 and 507 cm^{-1} were ascribed to the formation of GR, and the peak at 675 cm^{-1} was ascribed to the formation of magnetite [20,21], which indicated the co-existence of GRs and magnetite at 10 min of EC. At 20 and 30 min, only one peak at 675 cm^{-1} was observed, indicating that magnetite became the main crystalline phase of the formed flocs with the unstable GRs quickly transformed into magnetite in Fe(0) EC. As presented in Fig. 2B, the flocs formed at 10, 20 and 30 min at high DO concentration exhibited two bands centered at 233 and 366 cm^{-1} , which corresponded to the Fe–O vibration bands of Fe(III) hydroxides. These observed peaks were consistent with the typical peaks of γ -FeOOH [20].

XRD patterns of EC precipitates were obtained. For the flocs generated at low DO concentration (Fig. 2C), distinctive patterns of magnetite were observed [25,26]. The diffraction patterns for EC precipitate show three peaks at 30.1°, 35.5° and 43.1°, corresponding to (220), (311) and (400) planes of magnetite (JCPDS No. 88-0866). The intensity of the diffraction peaks increased with reaction time increasing from 10 min to 20 min, and remained nearly constant from 20 min to 30 min, showing a higher crystallinity of the formed precipitates with longer electrolysis duration in the Fe(0) EC process. No distinctive patterns of other crystalline structures were observed. In contrast, for EC precipitate at high DO concentration, poorly crystallized precipitate was observed (Fig. 2D).

The morphologies of the precipitates formed in EC under high and low DO conditions (5 mmol/L Na_2SO_4 , initial pH 5.0 ± 0.1 , 5 mA/cm^2) at the reaction time of 20 min were observed with TEM and EDAX analysis (Fig. S3 in Supporting information). For the flocs formed at low DO concentration, cubic particles were formed (Fig. S3A), and the morphology was consistent with the morphology of magnetite reported in previous work [21]. The

average size of the particles was estimated to be around 30 nm. For the flocs formed under high DO condition, poorly crystallized particles with shapes of bars were formed during Fe(0) EC (Fig. S3B). The average diameter was estimated to be 15 nm and average length was estimated to be about 100 nm. Corresponding SAED results showed the crystalline and amorphous structures of flocs formed under different conditions and EDAX results showed the presence of Sb on the flocs.

GRs are Fe(II)–Fe(III) hydroxides, and they belong to the family of layered double hydroxides (LDHs). The structure of GRs is composed of positively charged brucite-like layers and negatively charged interlayer anions that are balancing the charge [27]. The high adsorption capacity, reducing ability and high reactivity has been reported elsewhere. However, the GRs are unstable and transforms readily into more stable forms through the oxidation by air and other oxidants. In EC process, the generated GR could be an important intermediate because of its high adsorption ability, and they can play a significant role in the removal of Sb(V) during the reaction process. Therefore, if the GRs can be generated more steadily during EC, the removal efficiency of Sb(V) could be further increased. On the other hand, if the removal efficiency of Sb(V) do increase with increasing stability of GRs, it could further improve that GRs are the key species for Sb(V) removal. Therefore, we tried to achieve the stable generation of GRs by preventing the oxidation of Fe(II) through adding sodium sulfite into the EC process. Sodium sulfite has long been used in water treatment as an oxygen scavenger agent in industry by reacting with dissolved oxygen in the solution.

In this work, different amount of sulfite was added into Fe(0) EC system to improve Sb(V) removal efficiency by removing oxygen during EC instead of using N_2 purging, and in these experiments, the concentration of DO is not controlled using N_2 or O_2 . The removal efficiency of Sb(V) with the addition of sulfite is presented in Fig. 3. The addition of 0.2–1.0 mmol/L sulfite can improve Sb(V) removal efficiency. However, with the concentration of Na_2SO_3 increased to 2.0 mmol/L, Sb removal efficiency decreased (Fig. 3A). Three reasons may account for this adverse effect. Firstly, the addition of sulfite has a buffering effect which will slow down the increase of pH during EC (Fig. 3B), and the formation of precipitates will be hindered. Secondly, the excessive sulfite could be oxidized

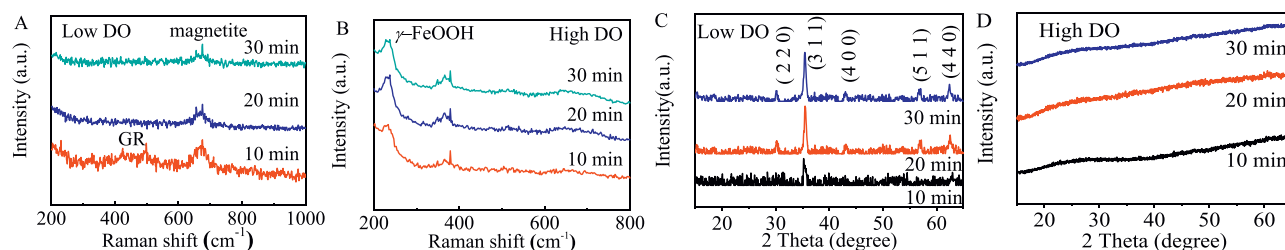


Fig. 2. Raman spectra of Fe(0) EC precipitates formed under low DO (A) and high DO (B) conditions; XRD patterns of Fe(0) EC precipitates formed under low DO (C) and high DO (D) conditions (5 mmol/L Na_2SO_4 , initial pH 5.0 ± 0.1 , 5 mA/cm^2).

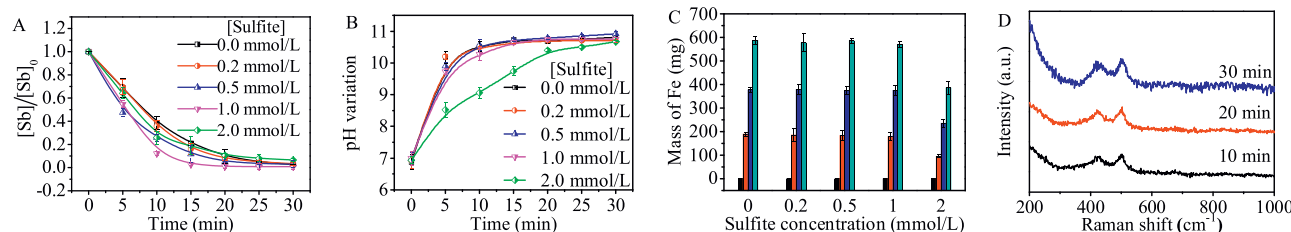


Fig. 3. (A) Sb(V) removal with the addition of Na_2SO_3 . (B) Variation of pH with the addition of different Na_2SO_3 concentrations. The mass of generated Fe (C) and Raman spectra (D) with the addition of 0.5 mmol/L Na_2SO_3 (5 mmol/L Na_2SO_4 , 5 mA/cm^2 , initial pH 5.0 ± 0.1).

on the anode, which competed with the anodic generation of Fe ions. This can be proved by the amount of generated Fe as presented in Fig. 3C, where a significant drop in the amount of generated Fe was observed with an initial concentration of sulfite of 2.0 mmol/L. Especially, at the initial concentration of sulfite of 2.0 mmol/L, the generation of Fe ions is very slow in the first several minutes, suggesting that sulfite was oxidized on the anode prior to the anodic generation of Fe from the anode. Thirdly, with excessive sulfite in the solution, the oxidation of Fe(III) was inhibited, and the formation of GRs was also inhibited since the formation of GRs requires partial oxidation of Fe(II) to Fe(III). With the existence of excessive sulfite, GRs cannot be effectively formed, which was unfavorable for Sb(V) during EC.

To understand the reason for the enhanced removal efficiency through sulfite addition, the formed precipitates with 1.0 mmol/L sulfite were characterized by Raman analysis. As shown in Fig. 3D, at 10 min, two peaks at 423 and 507 cm^{-1} were observed, indicating the formation GRs. Unlike the GRs formed in Fe(0) EC with N_2 purging, the precipitates formed with the addition of Na_2SO_3 were relatively stable even after 30 min of EC. Then the precipitates were stored underwater in a sealed centrifuge tube for two weeks, and then tested again. The peaks of GRs were still clearly observed, although a small part was transformed into $\gamma\text{-FeOOH}$. These results suggested that the GRs formed in Fe(0) EC with sulfite addition are quite stable even after two weeks.

As we have discussed above, GRs have relatively large specific surface area and high ion exchange capacity, and thus contributing to its ability in removing a lot of pollutants such as heavy metals and cations, etc. The addition of proper amounts of sulfite into Fe(0) EC system consumed both the DO in solution and the oxygen generated on the anode, which provided an anoxic environment and facilitated the generation of GRs. The improved Sb(V) removal efficiency with stabilized GR generation indicated that GRs played an important role in efficient Sb(V) removal in EC.

Phosphate is a commonly occurred anion in the environment, and it can react strongly with various iron oxides. Therefore, we investigated the influence of phosphate on the removal of Sb(V) under different phosphate concentrations (0–1.0 mmol/L). In these experiments, the concentration of DO was not controlled. Results are presented in Fig. 4A. When the phosphate concentration was relatively low (0.1 and 0.2 mmol/L), the influence of phosphate on the removal of Sb(V) in EC was relatively insignificant. Compared to the efficiency of Sb(V) in the absence of phosphate, in the initial stage of reaction (0–20 min), the removal efficiency of Sb(V) was slightly lower, but after 20 min, the removal efficiency increased and became slightly higher. With the increase of phosphate concentration (> 0.2 mmol/L), the inhibition effect began to dominate.

The reasons for these phenomena were then analyzed. Through the observation of the reaction process, it was found that the color of the generated flocs quickly turned into black in the absence of phosphate. But in the presence of phosphate, the green color can

maintain for a long time (> 30 min). This indicated that the presence of phosphate can actually affect the transformation of GRs in EC process, thus affecting the removal efficiency of Sb(V). Previous literature has reported a strong complexation effect between phosphate and iron oxides [28] and phosphate will adsorb strongly onto the surface of iron oxides. This will prevent the release of intercalated ions by forming a space barrier, and makes the GRs more stable and more resilient to oxidation. On the other hand, Sb(V) mainly exists as $\text{Sb}(\text{OH})_6^-$ anions, which can also adsorb onto the surface of iron oxides and is believed to be partially removed through adsorption [29] during EC in this work. In order to prove the effect of competitive adsorption of Sb(V) and PO_4^{3-} on the generated flocs, we also compared the removal of phosphate during EC in the presence and absence of Sb(V) (50 mg/L) and results are provided in Fig. 4B. In the presence of Sb(V), the removal of phosphate was significantly inhibited, indicating a possible competitive adsorption effect between PO_4^{3-} and Sb(V). Raman spectroscopies of the generated flocs were further acquired for the flocs formed with the presence of phosphate at different concentrations. In the presence of 0.1 mmol/L and 0.2 mmol/L phosphate, only peaks of GR were observed in the first stage of reaction, and magnetite was only observed at 30 min. When the concentration of phosphate was 0.5–1.0 mmol/L, only stable signals of GR were observed in the Raman spectra (Fig. 4C and Fig S4 in Supporting information). These results also indicated that the presence of phosphate can significantly increase the stability of GR during EC process.

The possible reasons for the influence of phosphate on Sb(V) removal during EC can be explained as follows: First, when the concentration of phosphate was low, the removal of Sb(V) was inhibited at the beginning of reaction because of the competitive adsorption with phosphate. The generation of GR was more stable when the phosphate was constantly removed. After the phosphate was consumed, the generated GR can promote the removal of Sb(V) in return, which made the removal of Sb(V) actually became higher. When the concentration of phosphate was high, the effect of competitive adsorption dominated during the whole reaction process and inhibited the removal of Sb(V) significantly. These results also indicated that GR was important in the removal of Sb(V) in EC process, and the stable generation of GRs could be very meaningful for pollutants removal in Fe(0) EC.

In conclusion, the relationship among Fe speciation, different reaction condition and Sb(V) removal efficiency in Fe(0) EC was studied in this work. It was found that the Fe species formed at low DO concentrations were mainly GR and magnetite, and $\gamma\text{-FeOOH}$ was identified to be the end product formed at high DO concentration. Sulfite and phosphate, if present in low concentrations, were proved to have the ability to stabilize the GR that had higher surface area and stronger adsorption ability for Sb(V), thus improving Sb(V) removal efficiency in EC. These results suggested the GR could be a key component for the efficient removal of Sb(V) in Fe(0) EC, and adding sulfite provided a simple way of increasing Sb(V) removal efficiency in Fe(0) EC.

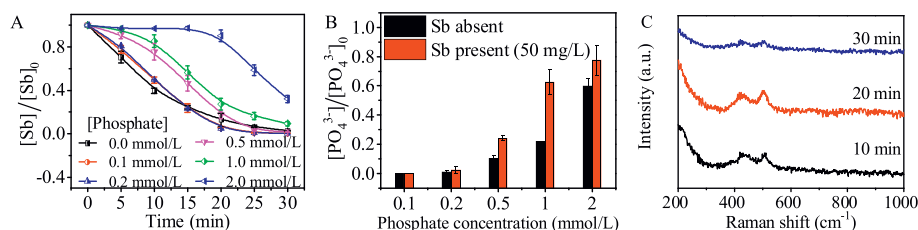


Fig. 4. (A) Sb(V) removal with the addition of different concentration of PO_4^{3-} . (B) The removal of PO_4^{3-} in EC in the absence and presence of 50 mg/L Sb(V) and (C) Raman spectra of the formed precipitates with the presence of 1.0 mmol/L phosphate (5 mmol/L Na_2SO_3 ; 5 mA/ cm^2 ; initial pH 5.0 ± 0.1).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.ccl.2020.07.024>.

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