# Removal of antimony from antimony mine flotation wastewater by electrocoagulation with aluminum electrodes

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**Abstract**

Antimony (Sb) has received increasing environmental concerns due to its potential toxic and carcinogenic properties. In the present work, the electrocoagulation technique was used to treat the flotation wastewater from a heavy antimony polluted area, and the mechanism of removing Sb was also investigated. The study focused on the effect of operation parameters such as current density, initial pH and standing time on the Sb removal efficiency. Antimony concentration of below 1 mg/L in the treated wastewater was achieved, which meets the emission standards established by State Department of Environmental Protection and State Administration of China for Quality Supervision and Inspection and Quarantine of China.

**Key words**: antimony; electrocoagulation; sodium butyl xanthate; aluminum

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## Introduction

Due to its potential or proven carcinogenicity, immuno-toxicity, genotoxicity, and reproductive toxicity, antimony (Sb) has been listed as the one of the prime pollutants by US EPA (1979) and Europe Union Environment Protection Agency (Council of the European Communities, 1976). Antimony can be introduced to the environment via various anthropogenic and natural processes such as automobile brake, antimony-containing medicines and wastewater from metallurgy, rubber and mining industries. Among those, the flotation wastewater (FW), which typically contains high concentrations of Sb in the presence of sodium butylxanthate and traces of arsenic (As) (Zhu et al., 2009), is a major pollution source of Sb. In general, FW comes from the Sb mineral processing where the flotation method is widely used to separate and concentrate Sb ores by altering their surfaces to a hydrophobic or hydrophilic condition. After the flotation, the processing wastewater is usually discharged into the tailing dam in the antimony mine, and some of the cleaned liquid is recycled to the flotation process. The leachate out of the tailing dam and the river around the antimony mining area contains high antimony contents (Zhu et al., 2009), as well as the surrounding soil and plants, which could have significant impacts on the environments.
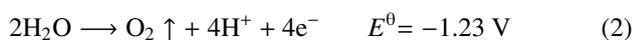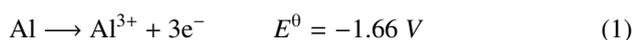
At present, only a few pretreatment methods to flotation wastewater have been reported such as adsorption (Imai et al., 1983; Bhattacharyya and Dutta, 1992; Hasany and Chaudhary, 1996; Khalid et al., 2000; Navarro and Alguacil, 2002; Nishiyama et al., 2003; Erosa and Höll, 2006; Xu et al., 2006), coagulation (Du, 1995; Nakamura and Tokunaga, 1996; Kang et al., 2003; Chen, 2005), membrane separation (Kang et al., 2000), solvent extraction (Navarro et al., 1999; Saleh et al., 2001), and electrochemistry (Koparal et al., 2004). Each has its advantages and limitations. The adsorption method is limited by the supply and added cost of absorbents such as rice hulls, river sands and titania (Bhattacharyya and Dutta, 1992; Hasany and Chaudhary, 1996; Khalid et al., 2000; Navarro and Alguacil, 2002). The coagulation technology is a commonly-used technology, which however has low removal efficiency of Sb (Kang et al., 2003). The membrane separation technology is effective to provide drinking water or high purity water at very low Sb concentration but comes with a high cost of membranes (Kang et al., 2000). The electrodeposition technology was once applied to

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electrorefining wastewater with high Sb concentrations up to 1500 mg/L (Koparal et al., 2004), but its application to wastewater of low Sb concentration has not been reported.

Electrocoagulation technology is an effective electro-chemical and water treatment method. It has gained increasing attention in recent years due to its simple operation, high removal efficiency, low energy consumption and low cost, as well as producing little sludge and without requiring too many chemicals (Holt et al., 2005; Mouedhen et al., 2008). The electrocoagulation technology had successfully applied to treat drinking water and municipal wastewater (Golder et al., 2007a, 2007b; Bagga et al., 2008; Heidmann and Calmano, 2008a; Mouedhen et al., 2008). The mechanism of electrocoagulation had been discussed in several previous publications (Emamjomeh and Sivakumar, 2006; Heidmann and Calmano, 2008a, 2008b), which include two main steps: (A) Aluminum anode dissolves and produces aluminum ions (Reaction (1)); oxygen gas is released from the anode as the increase of current density (Reaction (2)); and hydrogen gas is released from the cathode (Reaction (3)).
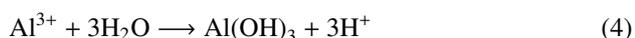
Anode:

$$Al \longrightarrow Al^{3+} + 3e^- \qquad E^\theta = -1.66\ V \qquad (1)$$

$$2H_2O \longrightarrow O_2 \uparrow + 4H^+ + 4e^- \qquad E^\theta = -1.23\ V \qquad (2)$$

Cathode:

$$2H_2O + 2e^- \longrightarrow H_2 \uparrow + 2OH^- \qquad E^\theta = -0.828\ V \qquad (3)$$

(B) $Al^{3+}$ immediately generates flocculated aggregates-hydrated alumina (FAHA) around anode and cathode (Reactions (4) and (5)).

Anode:

$$Al^{3+} + 3H_2O \longrightarrow Al(OH)_3 + 3H^+ \qquad (4)$$

Cathode:

$$Al^{3+} + OH^- \longrightarrow Al(OH)_3 \downarrow \qquad (5)$$

Because the produced FAHA can adsorb pollutants effectively, the hydrogen and/or oxygen gas can help flocculating particles floating in the solution, the electrocoagulation process could be a promising method to treat Sb-containing wastewater.

In FW, the main pollutants are antimony and sodium butyl xanthate (SBX) (Grandberg and Ou, 1994; Zhu et al., 2009). No study was available on the pretreatment of antimony-polluted FW by electrocoagulation technology. In the present work, treating the antimony-polluted flotation wastewater by electrocoagulation technology has been studied. Due to the existence of SBX, the removal mechanism of antimony may be different from that of without organic compounds. The removal mechanism of antimony and its influential factors, such as current density, pH and the treatment time, are also be investigated.

# 1 Experiments

## 1.1 Materials and analysis

Flotation wastewater (FW) sample was collected from Xikuangshan mine area in Hunan Province, Central China. The concentrations of total antimony and total arsenic were determined using a hydride generation-atomic fluorescence spectrometry (HG-AFS) (Jitian Instrument Ltd., China, detection limit is equal or lesser than 0.020 μg/L). The working conditions of HG-AFS are shown in Table 1. The stock solution of 1.0 mg/L total antimony was prepared from concentrated Sb (1000 mg/L from National Research Center for Certified Reference Material, China). The standard solution of total antimony was diluted progressively from the stock solution. The concentration of the SBX was determined using an ultraviolet-visible spectrophotometer (Puxi General Electric Co., Ltd., China). The most absorptive wavelength of sodium butyl xanthate was 301 nm. The elemental composition of the trace metals in the FW was obtained using a PE 5100 atomic absorption spectrophotometer (PerkinElmer Co., Ltd., USA, detection limit is 0.020 μg/mL). Electric conductivity of solution was determined with DDS-12A Digital Conductivity Meter (Pengshun Scientific Apparatus Co., Ltd., China). Concentrated hydrochloric acid, sodium borohydride (NaBH$_4$) powder, potassium iodide (KI) powder and ascorbic acid powder (vitamin C) are fine-chemical-grade regents. Concentrated sulfuric acid and solid sodium hydroxide are technical ultra-purified regents.

**Table 1** Operating conditions employed to determine antimony by HG-AFS

| Property | Value |
| --- | --- |
| Negative high voltage | 280 V |
| Atomizer hight | 8 mm |
| Lamp A current | 60 mA |
| Carrier gas | 300 mL/min |
| Dryer gas | 800 mL/min |
| Injection rate | 0.5 mL |
| Read time | 5 sec |
| Delay time | 1 sec |
| Measurement type | Standard curve |
| Signal type | Peak area |

The main physical and chemical characteristics of the FW are illustrated in Table 2. It clearly shows that Sb and SBX were the main pollutants, the concentration of As was low, and other metals such as Fe, Cu and Zn had not been detected. The FW has high conductivity (1.55 or 1.66 mS/cm), therefore it is not necessary to add electrolyte such as Na$_2$SO$_4$ or NaCl.

## 1.2 Apparatus and experimental procedures

The experimental equipment is an electrobath with aluminum electrodes in series (Fig. 1). The aluminum plates are the commercial grade aluminum sheet of 2 mm thickness (> 99%, Guiyang Aluminum Company, China) with a dimension of 130 mm × 40 mm. Two electrodes were placed 1 cm apart in a 500-mL beaker. The submerged surface area of each electrode was 18 cm$^2$.

**Table 2** Main physicochemical characteristics of flotation wastewater samples

| Sample | pH | Conductivity (mS/cm) | DO (mg/L) | K$^+$ (mg/L) | Na$^+$ (mg/L) | Ca$^{2+}$ (mg/L) | Mg$^{2+}$ (mg/L) | Total Sb (mg/L) | Total As (mg/L) | SBX (mg/L) |
|---|---|---|---|---|---|---|---|---|---|---|
| Sample 1 | 7.04 | 1.66 | 4.67 | 9.78 | 46.2 | 389 | 24.8 | 28.6 | 0.0246 | 0.376 |
| Sample 2 | 7.29 | 1.55 | 6.17 | 9.85 | 40.5 | 360 | 24.9 | 10.4 | 0.0104 | 0.434 |

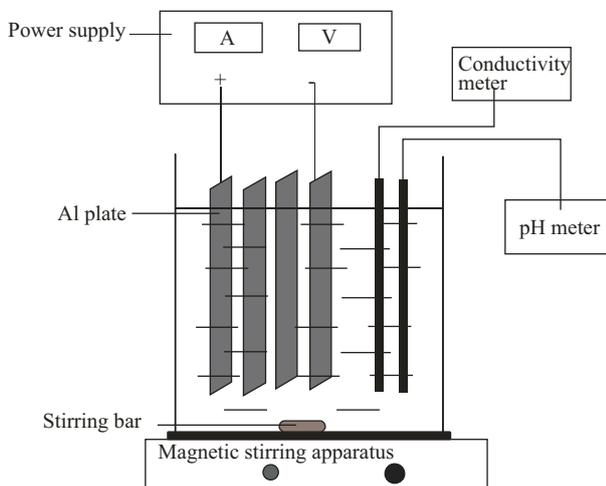BX: sodium butyl xanthate; Sample 1, 2: flotation wastewater samples.



**Fig. 1** Experimental apparatus for treating flotation wastewater by electrocoagulation.

The electrodes were abraded with sand paper to remove the oxide membrane and then cleaned with distilled water. A working volume of 500 mL was used. The mixing in the beaker was achieved magnetically. The initial pH value of the flotation wastewater was adjusted with NaOH or H$_2$SO$_4$ solution. The current was held constant for each run with electrolytic time of 60 min. Samples were taken from the reactor at proper time intervals during the course of experiments, and then centrifuged to separate the upper clear solution.

The direct current applied to the electrolytic cell was provided by a LW6J3 DC Regulator (Liyou Electric Co., Ltd., China). The electric conductivity of solution was determined by a DDS-12A digital conductivity meter (Pengshun Scientific Apparatus Co., Ltd., China). The stirring of solution was operated with a 78-L magnetic stirring apparatus (Pudong Physical and Optical Instrument Factory, China), and the pH of solution was determined with orion pH meter (Thermo Orion, USA).

## 2 Results and discussion

### 2.1 Antimony species in the flotation wastewater

The pH value, redox potential, and the presence of SBX and a flotation collector have significant influence on the antimony species in the FW. Granberg and Ou (1994) found antimony in the FW existed in forms of ([Sb(C$_4$H$_9$OCSS)$_3$]) and/or ([Sb(OH)$_6$]$^-$) (or SbO$_3^-$). The solution pH is an important factor to influence the speciation of antimony. When pH < 4, antimony existed in the form of trivalent antimony [Sb(C$_4$H$_9$OCSS)$_3$]. When pH > 9, [Sb(OH)$_6$]$^-$ is the main form of antimony. In the present

study, pH value of the flotation wastewater was 6–8, thus [Sb(C$_4$H$_9$OCSS)$_3$] and [Sb(OH)$_6$]$^-$ would be coexisted in the FW.

### 2.2 Effect of current density

Current density is one of the most important operating parameters in the electrocoagulation. The effects of current density on the removal of antimony and mass of flocculated aggregates are displayed in Fig. 2a, b, respectively. The antimony removal efficiency firstly increased with current density increasing from 55.5 to 166.7 A/m$^2$ and then reached a plateau when current density was up to 167 A/m$^2$. A similar phenomenon was also observed in that of As removal (Ratna et al., 2004). Same to the removal efficiency of antimony, the mass of flocculated aggregates was also increased at first and shown no obvious change at higher current density. Furthermore, a clear positive correlation was found between the antimony removal and the flocculated aggregate under the operational current density.

Because [Sb(C$_4$H$_9$OCSS)$_3$] and [Sb(OH)$_6$]$^-$ coexist in the solution, three reactions may occur at the anode during the electrocoagulation process, where aluminum anode dissolves first (Reaction (1)), oxygen is released from anode (Reaction (2)), and butyl xanthate ion is oxidized to dixanthogen (ROCSS)$_2$ on the anode (Reaction (6)):

$$2C_4H_9OCSS^- - 2e^- \longrightarrow (C_4H_9OCSS)_2 \quad E^\theta = -0.128 \text{ V} \tag{6}$$
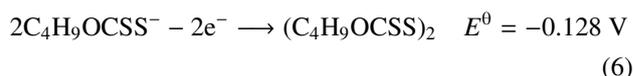
Due to the low concentration of SBX ion ($10^{-4}$ mol/L), based on the actual content of SBX in the flotation wastewater), the electrode potential of ROCSS$^-$/(ROCSS)$_2$ (0.109 V) and the evolution potential of oxygen gas
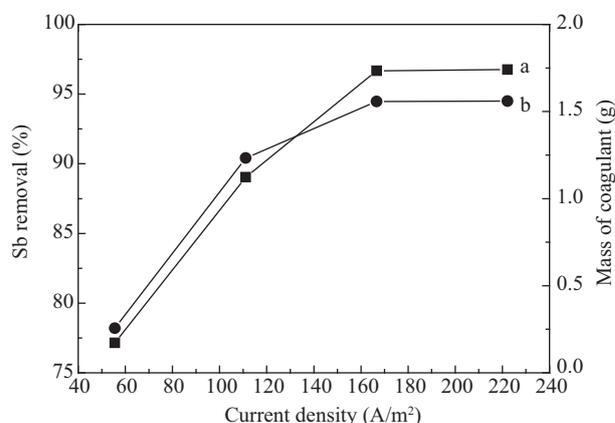


**Fig. 2** Effects of current density on the removal of antimony in the flotation wastewater (line a) and on the mass of coagulant (net weight) produced in the electrocoagulation process (line b). Condition: pH 2; anode surface 18 cm$^2$; electrolysis time 60 min.

(1.23 V) on aluminum electrode exceeded the electrode potential of aluminum (–1.662 V). Therefore, Reaction (1) would be the main reaction occurred at the anode and aluminum ions hydrolyzation resulted in a production of FAHA.

At the cathode, two reactions may occur: $H^+$ ions get electrons to produce $H_2$ (Reaction (3)) and $SbO_3^-$ ions are reduced to $SbO_2^-$ ions (Reaction (7)):

$$SbO_3^- + H_2O + 2e^- \longrightarrow SbO_2^- + 2OH^- \qquad E^\theta = -0.443 \text{ V} \tag{7}$$
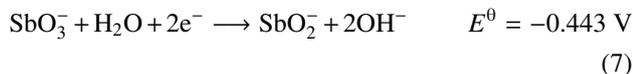
Super potential of hydrogen gas $\eta_{H2}$ the aluminum electrode rises with current density. Supposing that the concentrations of $SbO_3^-$ and $SbO_2^-$ are equal, the electrode potential of redox couple $SbO_3^-/SbO_2^-$ (–0.0286 V) is below the evolution potential of $H_2$, Reaction (3) should be the main reaction at the cathode.

It appears that the mechanism of removing antimony from the flotation wastewater is: (1) aluminum anode dissolves and produces FAHA firstly; (2) the flocculated aggregates adsorbs $[Sb(C_4H_9OCSS)_3]$ and $[Sb(OH)_6]^-$, and (3) $H_2$ bubbles produced at the cathodes float flocculated aggregates out of water. Similar mechanisms have also been reported by Heidmann and Calmano (2008a).

The applied current density has significant effects on the mass of flocculated aggregates. Faraday's law can be used to calculate mass ($m$) of flocculated aggregates:

$$m = \frac{ItM}{zF} \tag{8}$$

where, $I$ is the operating current density, $t$ is the runing time, $M$ is the molecule weight of aluminum hydroxide, $z$ is the number of electrons transferred in the anodic dissolution ($z = 3$), and $F$ is Faraday's constant. The equation shows that mass of flocculated aggregates is decided by the current density and run time. The mass of flocculated aggregates increased as current density increased at the first stage, but no obviously change can be found when the current density exceeding 166.7 A/m$^2$. It is speculated that aluminum plates were oxidized and deactivated from which aluminum plates blacked. Passivation of aluminum plates affected recycle of aluminum seriously.

## 2.3 Effect of initial pH

The pH value is another important factor in the electrocoagulation process. Effect of initial pH on removal efficiency of antimony is shown in Fig. 3. The removal efficiency of antimony reduced gradually with the increase in initial pH. The experimental data indicated that the mass of FAHA changed irregularly with initial pH and the removal rate of antimony had remarkable negative correlation with pH.

When the initial pH is 2, the solution is strongly acidic and the major antimony species is $[Sb(C_4H_9OCSS)_3]$. At the first stage of electrocoagulation process, aluminum anode dissolves (Reaction (1)) and hydrogen is released from cathode (Reaction (3)). $OH^-$ produced at the cathode are neutralized by $H^+$ (Reaction (9)):
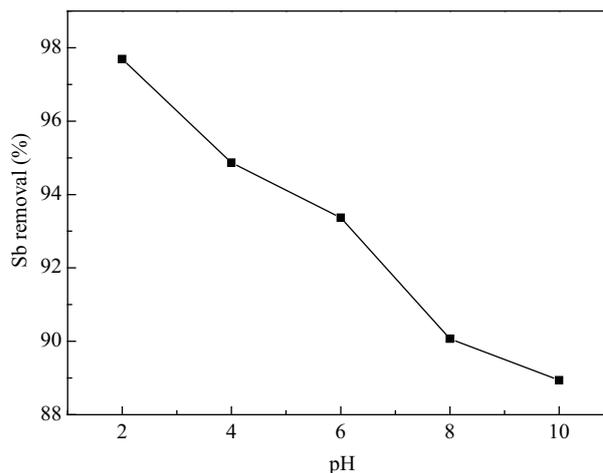
$$H^+ + OH^- \longrightarrow H_2O \tag{9}$$



**Fig. 3** Effect of pH on the removal of antimony. Condition: current density 166.67 A/m$^2$; anode surface 18.37 cm$^2$; electrolysis time 60 min.

Consequently, the mass of flocculated aggregates is small at the initial stage of electrocoagulation process (Reaction (5)). As the reaction (Reaction (9)) progresses, $H^+$ reduces with pH of the solution, $OH^-$ and mass of flocculated aggregates increases.

One possible explanation is that the removal efficiency of antimony reduced gradually with the increase in initial pH. Hydrogen ion ($H^+$) in the solution is smaller and $OH^-$ concentration is higher at higher pH values. However, due to the amphoteric characteristic of FAHA, some flocculated aggregates would dissolve based on the following Reaction (10):

$$Al(OH)_3 + OH^- \longrightarrow Al(OH)_4^- \tag{10}$$

The antimony adsorbed would be desorbed with some flocculated aggregates dissolving.

An alternative explanation is that, $[Sb(C_4H_9OCSS)_3]$ and $[Sb(OH)_6]^-$ coexists in the solution when pH is between 4 and 8; while $[Sb(OH)_6]^-$ was the main species of antimony when pH is up to 10. In general, FAHA adsorbs medium molecular more easily than negative ion because FAHA is negative in nature (Kang et al., 2003). Therefore, the removal efficiency of antimony at the initial pH of 4–6 is higher than that of the initial pH of 10.

However, the mass of flocculated aggregates changed irregularly with initial pH, for which the mechanism is still unclear, and further investigation is needed.

## 2.4 Effect of standing time

The removal efficiency of antimony did not change in 18 hr indicating that there is a strong attractive force between antimony and flocculated aggregates. It is widely believed that the predominant mechanism for the removal of soluble pollutant is interparticle bridging or coprecipitation in the coagulation process, and these actions may be the result of physisorption or chemsorption (Kang et al., 2003). In electrocoagulation process, coprecipitation may be the main mechanism to explain the removal of antimony from FW. Due to the presence of oxygen atom or hydroxyl group in the molecular structure of $[Sb(C_4H_9OCSS)_3]$ or $[Sb(OH)_6]$, forming hydrogen bond may be another major mechanism apart from the electrostatic attraction.

## 2.5  Settling velocity of flocculated aggregates

Setting velocity of flocculated aggregates affects the separation of upper clear solution and flocculated aggregates in the treated FW. The volume of flocculated aggregates with time in the settling process is shown in Fig. 4. It can be seen that volume of flocculated aggregates reduced gradually at the first stage, and then did not change after 5 hr. This phenomenon suggested that the optimum standing time of the treated FW should be determined for separating clear upper solution from flocculated aggregates.
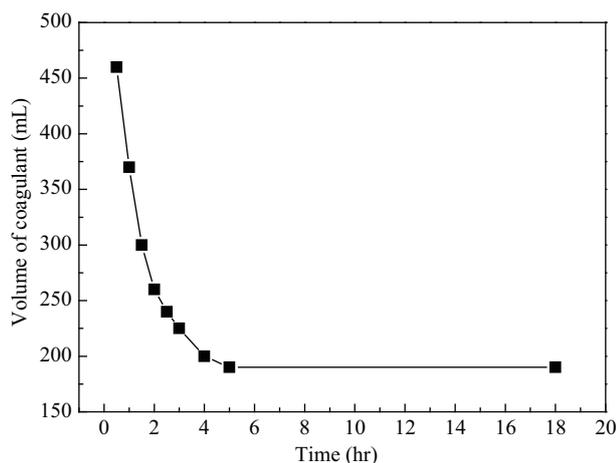


**Fig. 4**  Setting of coagulant during standing process.

## 2.6  Actual sample treatment by electrocoagulation

The results of treating FW by electrocoagulation are shown in Table 3. The removal efficiencies of antimony and arsenic were found to be independent of the initial concentration, which indicates that the electrocoagulation technology is an effective technology for treating FW. The removal efficiency of antimony up to 96% was obtained by electrocoagulation and the antimony concentration in the treated wastewater was below to 1 mg/L. The removal efficiency of arsenic up to 100% was obtained. Presently, the emission standard, established by state Department of Environmental Protection and State Administration for Quality Supervision and Inspection and Quarantine of China (exposure draft, 2008), is available for pollutants

**Table 3**  Results of flotation wastewater samples treatment by electrocoagulation

|  | Initial conc. (mg/L) | Final conc. (mg/L) | Removal rate (%) |
|---|---|---|---|
| **Sample 1** | | | |
| Sb | 28.60 | 0.83 | 97.7 |
| SBX | 0.3760 | 0.088 | 76.6 |
| As | 0.0246 | 0 | 100 |
| **Sample 2** | | | |
| Sb | 10.40 | 0.34 | 96.7 |
| SBX | 0.434 | 0.127 | 70.7 |
| As | 0.0104 | 0 | 100 |

SBX: sodium butyl xanthate.

from stannum, antimony, mercury industries. The residue level of antimony in treated wastewater (1 mg/L) is much less than the emission limitation of antimony (3 mg/L), indicating the electrocoagulation is a promising technology for antimony removal from the industrial wastewater. While, the removal rate of SBX was not high, needed to be treated further.

## 3  Conclusions

In the present work, the removal mechanism of antimony in the FW from antimony mine by electrocoagulation technique with aluminum electrodes has been investigated. Effects of impact factors such as current density, initial pH and standing time on the removal of antimony were studied. Due to the presence of sodium butyl xanthate , the removal mechanism of antimony is different from other metals. The result showed that the mass of flocculated aggregates and removal efficiency of antimony increased with the increase in electric current. The removal rate of antimony was positively related to the current density and mass of flocculated aggregates. The initial pH of solution affected the existing forms of antimony and negatively correlated with the removal rate of antimony. FAHA adsorb medium molecular more easily than negative ion because FAHA is negative in nature. Under the experimental conditions, the removal efficiencies of antimony in FW samples up to 96% were achieved, independent of the initial pH value.

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