Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2019.23740

150 (2019) 166–191
May

Combination of electrokinetics and nano zero-valent iron-based adsorption enhances Sb(V) removal from feed water in the batch and column mode processes

Tao Huang[a,*], Longfei Liu[a,*], Jiaojiao Xu[a], Shuwen Zhang[b,c]

[a]*School of Chemistry and Materials Engineering, Changshu Institute of Technology, 215500, China, emails: ht1104705720@qq.com (T. Huang), longfeiliu_cslg@163.com (L. Liu), 1037495839@qq.com (J. Xu)*
[b]*State Key Laboratory for coal mine disaster dynamics and control, Chongqing University, 400044, China, email: tillg14536@yahoo.com (S. Zhang)*
[c]*School of Resource and Environmental Science, Chongqing University, Chongqing, 400044, China*

Received 19 June 2018; Accepted 1 January 2019

ABSTRACT

Disorder mining and massive consumption of Sb-bearing ores have caused a dramatic rise in the accumulation of Sb related pollution in China over the past decade. The combination of iron-based adsorption and electrokinetics was applied in the batch and column experiments to enhance removal of Sb(V) anions from the feed water. Activated carbon supported nano zero-valent iron (AC-nZVI) was prepared as the filling adsorbent for the adsorption experiments. The maximum monolayer-adsorption capacity of AC-nZVI was 20.49 mg/g according to the Langmuir isotherm results. Sufficient dose of adsorbent with sufficient dose and pH ranging from 4 to 5 were favored in the equilibrium adsorption. Electrokinetics clearly had enhanced Sb(V) removal from the feed water with the maximum removal efficiency of 96.42% being achieved in the coupling system. The variable changes significantly affected the removal results of Sb(V) ions in the coupling experiments at a batch mode. The antimony species can be effectively removed from the stock solution in the continuous process. The flow rate of 40 mL/min was favored in facilitating the excellent performance of the assembled units at a minimum operating cost. The adsorbent regenerated within three cycles is still reliable in terms of removing Sb in a continuous treatment process.

Keywords: Nano zero-valent iron; Electrokinetics; Sb(V) removal; Activated carbon; Continuous process

1. Introduction

The guideline value of Sb in drinking water recommended by World Health Organization (WHO) is 10 μg/L [1]. Minister of Health of China (MOHC) even lowers the limited concentration of Sb in drinking water to below 5 μg/L (GB3838–2002). Sb can be gradually accumulated in the organs and tissues going through the respiratory and digestive systems (i.e., Bioaccumulation). Excessive intake of Sb can cause some symptoms, such as vomiting, liver cirrhosis, nephritis, and muscle necrosis [2,3]. The high concentration of Sb in the soils also can adversely retard the growth of vegetables, inhibit the photosynthesis, and disrupt the metabolism of plants. Sb exists in four chemical valences including Sb(III), Sb(0), Sb(III), and Sb(V) [4,5]. The compounds of Sb(III) and Sb(V) are more typically observed in the geochemical system in comparison with the other two states. Inorganic compounds of Sb (e.g., inorganic antimonite and antimonate) have higher toxicity than its organic counterparts (e.g., methylated species) [6,7]. The harmfulness of Sb species for human health and ecological system follows the order of Sb > Sb(III) > Sb(V) > organoantimonials. Although, Sb(III) species are estimated

---

* Corresponding authors.

to be approximately 10-fold more toxic in contrast to Sb(V) oxyanions, the removal for Sb(III) is more effective compared with Sb(V) using the different coagulants and adsorbents [8]. In comparison with Sb(III), Sb(V) compounds are more stable and more predominant in the mildly reducing and aerobically aquatic conditions. Sb(III) is subjected to the oxidation at more negative redox potential (E_h) values. Therefore, the efficient removal of Sb(V) species is a key factor in purifying and remediating Sb-contaminated wastewater [9].

The positively charged Sb(V) species in an inorganic form (i.e. SbO_2^+) is only observed in a very acidic environment (i.e., pH < 2). Whereas, a negatively charged Sb(V) form (i.e., $H_2SbO_4^-$, $Sb(OH)_6^-$) is most commonly recognized over a wide pH range of 2–12. Many techniques have been used to remove Sb(V) (oxy)anions from the wastewater, although the total numbers of relevant publications are limited [10,11]. The frequently mentioned technologies include coagulation/flocculation, adsorption, ion-exchange, (bio)filtration (e.g., microfiltration, ultrafiltration, nanofiltration), (bio) electrochemical methods, phytoremediation, etc. [3,7,12]. The adsorption has comparatively attracted more attention due to its intrinsic advantages, such as procedure-simplicity, cost-effectiveness, and minimal secondary-pollution. A variety of adsorbents including activated carbons, clay minerals, and metal oxides have been proposed and applied to remove Sb species from the contaminated water in the vicinity of the Sb involved industries [13–15]. However, some iron minerals (e.g., goethite, hematite, and ferrihydrite) existing in the soils are preferably used to immobilize Sb species and to reduce the leachability of pollutants during the naturally occurring processes due to their abundance and less costs.

Several studies on preparing iron hydroxides and oxyhydroxides (natural or chemically modified) for the adsorption of Sb(III) and Sb(V) ions from water on a bench scale have been reported. Although, some acceptable results have been achieved, the maximum adsorption capacities of iron adsorbents referred in the previous studies are still low and the recycling generation of adsorbents and the continuity of treatment process for purifying Sb-contaminated water have been rarely investigated [6,16,17]. Considering that electrokinetics (EK) is a compatible method [18–20], commonly combining with other technologies to enhance the remediation of solid wastes and wastewater by several mechanisms including hydrolysis, electromigration, electroosmosis, and electrophoresis [21–23]. To overcome the drawbacks caused by the iron-based adsorbents, EK was incorporated into a traditional iron-based adsorption process in this research. A synergetic process of nano zero-valent iron-adsorption and EK in a column form was assembled to remove Sb(V) anions from the stocking solution (i.e., feed water). The EK was used to strengthen transfer of Sb(V) in the electrolyte. The homemade activated carbon (AC) of *Neosinocalamus affinis* bamboo was prepared for the loading of nano zero-valent iron (nZVI) to avoid the agglomeration of nZVI during the removal experiments. Objectives of this study were: (1) to study the effects of the initial pH and adsorbent dose on the removal efficiencies of Sb(V) in the equilibrium adsorption tests and to evaluate the maximum adsorption capacities (Q_m, mg/g) of adsorbent; (2) to quantitatively determine the optimization combination of the operating parameters including the initial pH, disposal time, Sb(V) concentration and voltage gradient in a single treatment unit; (3) to analyze the effects of the competing ions on the adsorption of Sb(V) during treatment process; (4) to evaluate an appropriate flow rate of the feed water for maximizing the treatment capacity of the processing unit; (5) to recycle and regenerate the adsorbents and to explore the regeneration capability of the continuous process.

2. Methods and materials

2.1. Chemical reagents

All chemical reagents referred in the research were purchased from SINOPHARM GROUP Co., Ltd. (Shanghai, China) and Sigma-Aldrich (USA), and were used without any further purification. The chemicals include ethanol (95%, AR), $NaBH_4$ (99.5%, AR), $K_2HPO_4 \cdot 3H_2O$ (99%, AR), humic acid (≥90%, FA), $FeCl_3 \cdot 6H_2O$, $FeSO_4 \cdot 7H_2O$, H_2SO_4 (98%), NaOH (97%, GR), Na_2CO_3 (≥99.8%, GR), Na_2SiO_3 (≥98%, AR) and $KSbO_6H_6$ (99.0%, AR), respectively. The deionized (DI) water obtained from a DI generator (panike, Hugke) and the solutions used for the preparation of nZVI were further degassed by passing a constant argon inflow. All the experimental vessels were immersed in a diluted HNO_3 solution (i.e. 15%) and continuously rinsed three times with DI water before the tests.

2.2. Preparation of stock solution and adsorbent

A standard Sb(V)-stock solution (200 mg/L) was prepared by dissolving $KSbO_6H_6$ in DI water. The feed water with different concentrations of Sb(V) ions was obtained by diluting the standard solutions with DI water. The initial acidity or alkalinity in feed water was adjusted by titration of 1.0 M H_2SO_4 or 1.0 M NaOH, respectively. The adsorbent was made by chemically loading nZVI on the homemade activation carbons (AC). The preparation of biomass-based AC (i.e. *Neosinocalamus affinis* bamboo as the raw precursor) was specifically detailed in the previous research [24,25]. AC supported nZVI (AC-nZVI) was synthesized through the wet impregnation and subsequent $NaBH_4$ reduction. The ferric ethanol was prepared by mixing $FeCl_3 \cdot 6H_2O$, and ethanol. The AC materials were soaked in 1 M ferric ethanol at the ratio of 1:20 (*m*/*v*, g/mL) over 1 h, which was further heated to evaporate the solvent and then dispersed in 100 mL fresh ethanol. Furthermore, 0.5 M $NaBH_4$ was added drop-wise to the mixture to achieve the reduction of ferric ions under nitrogen (N_2) atmosphere. The final AC-nZVI was filtered and rinsed with pure ethanol three times and dried at 80°C in vacuum.

2.3. Aqueous equilibrium-adsorption tests

0.1, 0.2, 0.3, 0.4, and 0.5 g of AC-nZVI granules were added to 100 mL of Sb(V) stock solutions (100 mg/L) respectively, to analyze the effect of adsorbent dosage on the removal of Sb(V) in some conical flasks. The reaction mixtures were shaken on a horizontal oscillator (KJDHZ-82, Zhonghui) at 120 rpm and 25 °C for 30 min. The influence of pH on the equilibrium adsorption of homemade adsorbent was evaluated in the range of 4.0–10. Sb(V)-stock solutions with different concentrations (e.g., 30, 60, 90, 120, 150, and 180 mg/L) were prepared by diluting the standard solutions using DI water. Three models used for isotherm adsorption were adopted to evaluate Q_m (mg/g) of adsorbent and were specified in the supporting information (Eqs. (S1)–(S3)).

2.4. Adsorption of Sb(V) in the batch

For Sb(V) adsorption, some experiments were conducted in a single EK-treatment unit in the batch. The schematic cross-section of cylindrical device used for EK experiments is shown in the supporting information (Fig. S1). Carbon clothes (WOS1002, i.e., Hydrophilic) at the top and bottom of apparatus were connected to a direct-current (DC) power source (MCH-K305D, 30V 10A) by wires, known as anode and cathode, respectively. The homemade adsorbent was closely packed in the cylindrical container (polycarbonate, height: 10 cm, diameter: 10 cm) for removal experiments. The feed water was continuously injected into the treatment unit through an inlet using a peristaltic pump (Lab 2015, SEHNCHEN) at 100 mL/min before turning on the power source. The blowholes distributing on the top of device lid were used to disperse O_2 and H_2, generating at the surfaces of anode and cathode, respectively. The effluent used for the concentration detection of antimonate was collected from the reservoir after EK process. To comprehensively, study the effects of the operating parameters on the treatment performance of the unit device, four factors were adjusted to conduct the orthogonal tests (Table 1). Four factors at four different levels in the orthogonal tests are specifically defined in Table S1 in the supporting information. The anions including HPO_4^{2-}, CO_3^{2-}, and SiO_3^{2-} all at the concentrations of 500 mg/L were added to the stock solution and were used to explore the interference of these competing ions on the adsorption of Sb(V) in an EK system.

2.5. Adsorption of Sb(V) in the column

Three treatment units were connected in serials, serving for the adsorption experiments in a column mode. The schematic diagram of assembled device is shown in Fig. 1. Filter cloths (Φ 25 μm) were fixed at the ends of each cylindrical unit in order to block some tiny adsorbent particulates. The flow rate of feed water was adjusted by a peristaltic pump, ranging from 20 to 100 mL/min, to explore the effects of flows on the removal of Sb(V) anions, treatment capacity (L), and operating costs over time. pH changes and redox potentials (ORP, E_h) were recorded throughout the disposal process at an interval of 5 min using a high-precision pH meter (BPP-920, BELL, USA) for t-test analysis. The experiments would be suspended once the concentrations of Sb(V) just arrived at 5 ug/L. The adsorbent was warily collected from the experimental apparatus after adsorption experiments. AC-nZVI particles unloaded from EK system were recycled by mixing with 1.0 M H_2SO_4 in a beaker, magnetically stirring at 1,200 rpm for 30 min, separating by a separator, and rinsing three times using DI water. The adsorbent was then reloaded to the treatment units to investigate the regeneration capability of column system.

2.6. Analysis methods and calculations

The zeta potentials of AC and AC-nZVI were measured to determine the point of zero charges (pH_{zpc}) by a micro-electrophoresis apparatus (JS94H, Zhong Chen Digital Technology Equipment Co., Ltd.) with the conductive solution of 0.1 M NaCl. The concentration of Sb(V) in the solution was measured in triplicate readings by ICP-MS (7500a, Agilent, USA). The removal efficiency of Sb(V) (i.e., $Re_{Sb(v)}$) in the effluent was calculated following Eq. (1), where C_0 is the initial concentration of antimonate, and C_t represents the concentration of Sb(V) after the experiments. The optimal combination of parameters in terms of batch-mode

Table 1
Design of orthogonal tests and the corresponding experimental results (Four factors with four levels, ($L_{16}(4^5)$))

Test No.	A[a]	B[b]	C[c]	D[d]	Removal efficiency (%)
1	1 (0 V/cm)	1 (5 min)	1 (4)	1 (50 mg/L)	78.84
2	3 (1.0 V/cm)	3 (15 min)	1	3 (150 mg/L)	93.32
3	4 (1.5 V/cm)	4 (20 min)	1	4 (200 mg/L)	89.17
4	2 (0.5 V/cm)	2 (10 min)	1	2 (100 mg/L)	90.55
5	2	4	3 (8)	1	95.87
6	4	3	2 (6)	1	94.48
7	3	2	4 (10)	1	92.28
8	1	4	4	3	76.24
9	4	1	4	2	84.63
10	1	3	3	2	80.89
11	2	3	4	4	82.21
12	2	1	2	3	79.92
13	3	1	3	4	85.21
14	3	4	2	2	96.42
15	4	2	3	3	88.14
16	1	2	2	4	73.23

[a]Voltage gradient (V/cm); [b]Proposing time (min); [c]Initial pH; [d]Sb(V) concentration (mg/L).

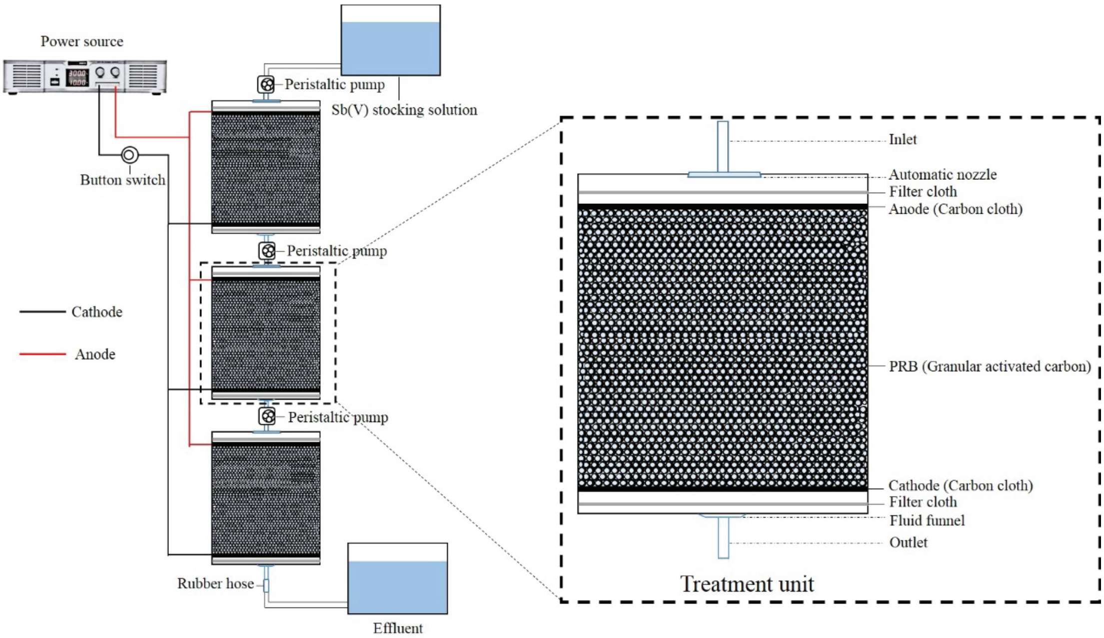

Fig. 1. Schematic diagram of the assembled device.

adsorption experiments was quantitatively evaluated based on the significance analysis and marginal means (Tukey's test, default $\alpha = 0.05$) of factors. The significance probabilities calculated by the sequential calculations of the type III sum of squares (SS), mean squares (MS), and F values (F) were used to directly indicate the significance of each factor on the target (i.e, $\text{Re}_{\text{Sb(V)}}$). The t-test analysis on pH and redox was conducted according to the comparison of average values between the different experimental groups. To quantify the operating costs and to evaluate the economic feasibility of the coupling technology for a further industrially continuous treatment of Sb(V)-contaminated wastewater, the cost (C_{op}, \$/m^3) was assessed by Eq. (2), where S_1 is a commercially evaluated price of AC-nZVI per ton (\$/t), S_2 is an international price of electricity per kW·h (\$·kW^{-1}·h^{-1}), m is the mass of the adsorbent (t), n is the cycling times of activated carbon, V_{electric} is the voltage (V), I is the direct current through the circuit (A), and V_{effluent} represents volute of the treated water (m^3).

$$\text{Re}_{\text{Sb}(V)} = \left(1 - \frac{c_t}{c_0}\right) \times 100\% \tag{1}$$

$$C_{\text{op}} = \frac{m \times \dfrac{S_1}{1 + 0.5n} + S_2 \times V_{\text{electric}} \times \int_0^t \text{Idt} \times 10^{-3}}{V_{\text{effluent}}} \tag{2}$$

3. Results and discussion

3.1. Equilibrium adsorption of Sb(V) by AC-nZVI

The solution pH not only affects the chemical speciation of Sb(V) existing in the aqueous environment, also dramatically changes the adsorption preference of adsorbent for contaminants. The pH_{zpc} values of AC and AC-nZVI were approximately 2.77 and 4.79, respectively, based on the pH values detected at the zero of zeta potential (Fig. 2(a)). The surfaces of adsorbents are positively charged when pH in the solution is lower than the corresponding pH_{zpc} values, which would facilitate the adsorption of negatively charged Sb(V) anions. The adsorption capacities (mg/g) of AC-nZVI decreased with the initial solution pH increasing from 4 to 10, regardless of the changes of adsorbent dosages (Fig. 2(b)). The maximum adsorption capacities (mg/g) of AC-nZVI were all achieved at pH ranging from 4 to 5 for each dosage. The equilibrium-adsorption results (Fig. 2(b)) of AC-nZVI echoed the results of pH_{zpc} values (Fig. 2(a)), indicating the electrostatic attraction indirectly changed by the solution pH, which had a prominent influence on the adsorption or removal of Sb(V) anions (i.e., $\text{H}_2\text{SbO}_4^-$and Sb(OH)_4^- from the stock solution. The adsorption capacity of AC-nZVI was always highest at the dosage of 0.5 g in either pH condition. The adsorption capacities of AC-nZVI granules increased with the increase of adsorbent dosage. The results demonstrated that the proposal of continuous treatment using AC-nZVI adsorbents for Sb(V)-contaminated solution was technically feasible. The adsorption experiments controlled by AC materials were also conducted under the same conditions to directly compare the adsorption capacities of Sb(V) between AC and AC-nZVI (Fig. 2(c)). The adsorption capacities (mg/g) of AC clearly decreased over the elevated pH values. The lower pH (i.e., pH < 4.0) was favored in removing Sb(V) anions using AC granules. The increased dosage also elevated Sb(V) adsorption of AC as same as that of AC-nZVI. Comparatively, the adsorption capacities of AC-nZVI (Fig. 2(b), i.e., 15–17 mg/g) were larger than the counterpart of AC (Fig. 2(c), i.e., 6–7 mg/g) at either same pH or either same dosage, indicating the load of nZVI on the AC had significantly strengthened the adsorption capacity of the AC materials for antimonate. More active sites can be surely presumed on the surfaces of AC-nZVI during the equilibrium adsorption process.

3.2. Evaluation of the maximum adsorption capacity

The potentiality of AC-nZVI in adsorbing Sb(V) anions from the stock solution should be clarified before the coupling experiments, which is essential for evaluating the performance of AC-nZVI in the electrolyzer. The affinity of AC-nZVI for Sb(V) can be quantitatively characterized by the evaluation of Q_m values, which are commonly obtained by the model fittings of experimental data. The fitted curves of Langmuir and D-R for the adsorption of AC and AC-nZVI under different concentrations of Sb(V) are shown in Fig. 3. The corresponding isotherm parameters are listed in Table 2. The adjusted correlation-coefficient values (Adj. R^2) showed that the adsorption of Sb(V) using both AC and AC-nZVI followed Langmuir isotherm rather than D-R model. Furthermore, the fitted curves visually verified the appropriateness in analyzing the adsorptive behaviors using Langmuir isotherm. The fitting results indicated that the removal or adsorption of Sb(V) anions took place through a mono-layer adsorption onto the surfaces of AC and AC-nZVI. As shown in Table 2, the maximum monolayer adsorption capacity (q_0, mg·g^{-1}) of AC-nZVI was approximately 20.49, which was higher than 10.27 of AC and 18.92 of La-doped magnetic biochar [26] while lower than 250.3 of Fe-Mn binary oxides [27]. The experimental results demonstrated that the loading of nZVI on AC particles had remarkably increased the adsorption capacity of AC while had not changed the adsorption pathway of AC-based adsorbents.

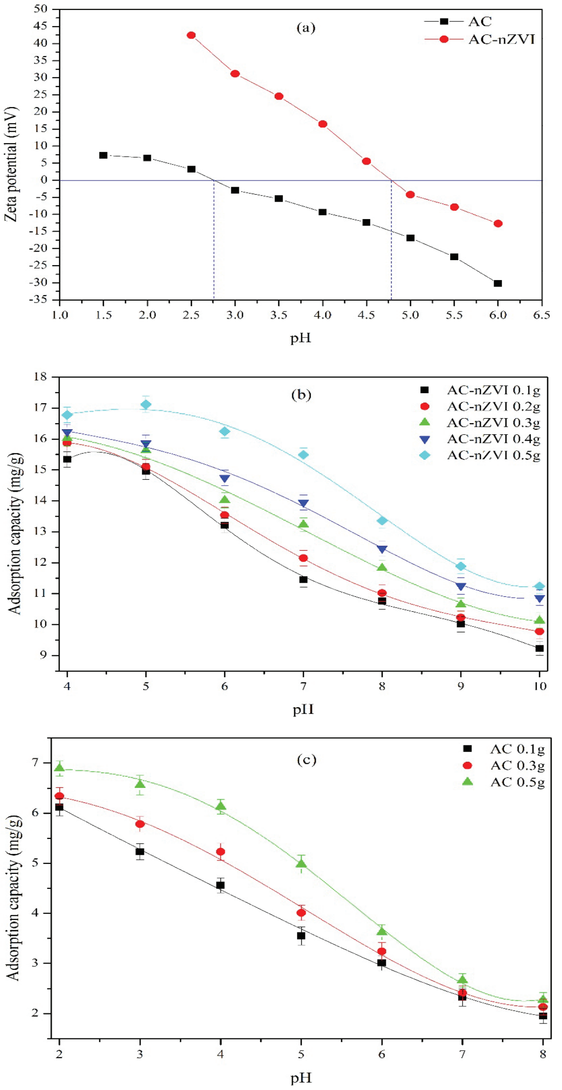

Fig. 2. Equilibrium adsorption of antimonate: zeta potentials of AC and AC-nZVI over pH (a), effect of initial pH and dosage using AC-nZVI (b) and AC (c).

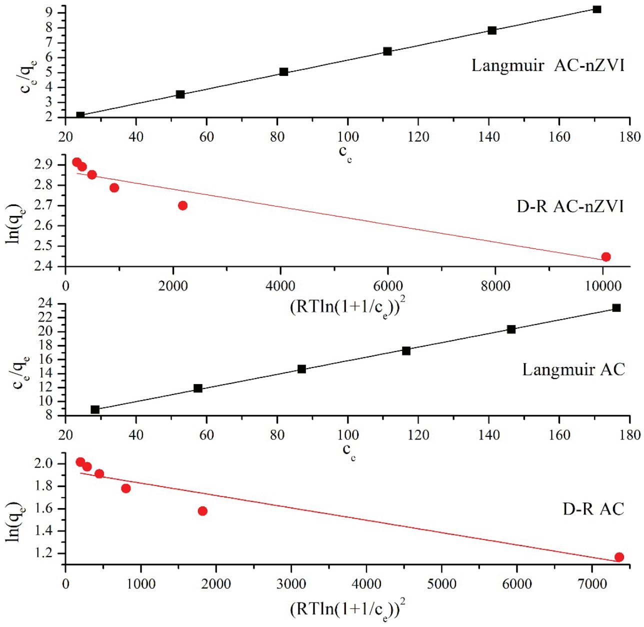

Fig. 3. Fitted curves of isothermal models, Langmuir and Dubinin-Radushkevich.

Table 2
Isotherm parameters of Sb(V) adsorption using AC and AC-nZVI (at initial pH of 4, adsorbent dosage of 0.1 g and 25°C)

Models	Parameters	AC	AC-nZVI
Langmuir $\frac{c_e}{q_e}=\frac{1}{q_0k_l}+\frac{c_e}{q_0}$	k_l	0.01592	0.05039
	q_0	10.26799	20.49180
	Adj. R^2	0.99899	0.99961
Dubinin-Radushkevich $\ln(q_e)=\ln(q_m)-B\varepsilon^2$	q_m	6.95833	17.59509
	B	1.11E–4	4.35E–5
	Adj. R^2	0.88741	0.90285

3.3. Optimization of the coupling system at a batch scale

3.3.1. Significance analysis of factors

In contrast to the adsorption in the chemical system, the adsorptive behavior of an adsorbent in the electrochemical system is not only dependent on the chemical form and the initial concertation of adsorbate also highly affected by the EK mechanisms. The anions and cations can be migrated to the electrodes through electromigration in the electrolyzer, which can further influence the diffusion of Sb(V) in the interface. As known, the water molecules are continuously decomposed on the anode (Eq. (3)) and cathode (Eq. (4)), strongly changing pH distribution in the electrolyte. To fully optimize the adsorption process in the synergistic treatment unit, the voltage gradients between anode and cathode should be cautiously selected. As shown in Table 1, the maximum removal of 96.42% by AC-nZVI in the unit was obtained in the combination of $A_3B_4C_2D_2$ at No.14 test (i.e., voltage gradient of 1.0 V/cm, proposing time of 20 min, initial pH of 6, and Sb(V) concentration of 100 mg/L). The significance analysis and marginal means based on $Re_{Sb(V)}$ values for each factor in the batch-mode experiments (default $\alpha = 0.05$) is shown in Table 3 and Fig. 4, respectively. The effects of variable interactions on Sb(V) removal are analyzed and listed in supporting information (Tables S2–S12). The significance probabilities of four parameters were 0.001, 0.008, 0.035, and 0.006, respectively, all lesser than the default α. This means the null hypothesis of insignificance should be denied, the changes of four factors (within four levels) all had significant effects on Sb(V) removal. An appropriate adjustment of the factors before the treatment process for each factor is vital to achieving desirable aims.

$$\text{Anode}: 2H_2O - 4e^- \rightarrow 4H^+ + O_2\uparrow \tag{3}$$

$$\text{Cathode}: 4H_2O + 4e^- \rightarrow 4OH^- + H_2\uparrow \tag{4}$$

3.3.2. Optimization of the treatment unit

As shown in Fig. 4, the $Re_{Sb(V)}$ values of 77.30%, 87.14%, 91.81%, and 89.11% were obtained at the voltage gradients (V/cm) of 0, 0.5, 1.0, and 1.5, respectively. The results indicated that EK treatment obviously enhanced Sb(V) removal from feed water. The voltage gradient of 1.0 V/cm made the treatment unit achieve the best performance among four levels. Meanwhile, $Re_{Sb(V)}$ increased from 82.15% to 89.43% with a gradual increase of the proposing time from 5 to 20 min, meaning a properly longer time setting was favored during the process. $Re_{Sb(V)}$ values of 87.97%, 86.01%, 87.53%, and 83.84% were got at the initial pH of 4, 6, 8, and 10, respectively, indicating the strong alkalinity should be explicitly avoided in the solution before being fed to the processing cell. $Re_{Sb(V)}$ progressively decreased from 90.37% to 82.465% with the elevation of the initial concentration of Sb(V) in the feed water. Overall, the optimal combination for the four parameters was determined as $A_3B_4C_1D_1$ including the voltage gradient of 1.0 V/cm, proposing time of 20 min, initial pH of 4, and Sb(V) concentration of 50 mg/L, which guaranteed the achievement of Sb(V) removal in a treatment unit.

3.4. Effect of competing ions on the adsorption of Sb(V)

To explore the potential effect of competing anions on the removal efficiencies of Sb(V) in the coupling system, the competing experiments at a batch mode were conducted at the voltage gradient of 1.0 V/cm, proposing time of 20 min,

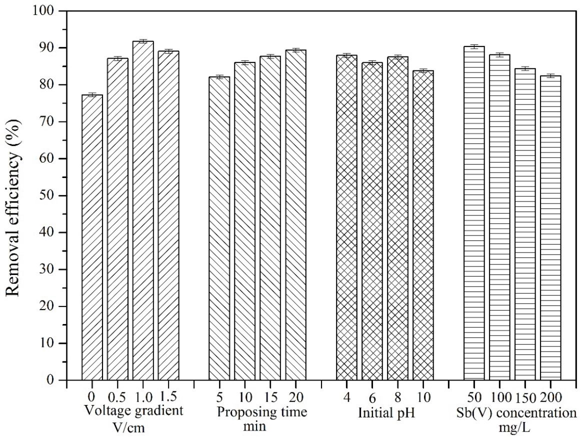

Fig. 4. Marginal means of the operating parameters in the coupling system.

Table 3
Significance analysis with the dependent variable of Sb(V)-removal efficiency ($Re_{Sb(V)}$, %) in the batch-mode experiments (default $\alpha = 0.05$)

Source	Type III sum of squares	df	Mean square	*F*	Sig.
Corrected model	790.527[a]	12	65.877	57.357	0.003
Intercept	119266.623	1	119266.623	103841.037	0.000
Voltage gradient	479.585	3	159.862	139.186	0.001
Proposing time	116.303	3	38.768	33.753	0.008
Initial pH	41.697	3	13.899	12.101	0.035
Sb(V) concentration	152.942	3	50.981	44.387	0.006
Error	3.446	3	1.149		
Total	120060.595	16			
Corrected total	793.973	15			

[a] *R* Squared = 0.996 (Adjusted *R* Squared = 0.978).

and initial pH of 4. The removal results obtained from feed water with different concentrations of 50–200 mg/L are shown in Fig. 5. Sb(V) removal was obviously affected by the presence of phosphate in the feed water. However, $Re_{Sb(V)}$ values increased from 42.58% to 61.53% step by step with the elevation of initial Sb(V) concentrations. The results indicated that the interference of phosphate (500 mg/L) on the adsorption of Sb(V) in the treatment unit can be moderately relieved by increasing the initial Sb(V) concentrations. The removal efficiencies ranging from 80.02% to 83.56% were obtained for the group of CO_3^{2-}. It seems the carbonate had an insignificant or minor influence on inhibiting Sb(V) removal during EK process compared with the other two groups. The $Re_{Sb(V)}$ values affected by SiO_3^{2-} were 28.62%, 34.19%, 36.15%, and 40.94%, respectively. Although, the increase of the initial Sb(V) concentration in the feed water slightly eased the interference of SiO_3^{2-}, the most significant inhibition brought by SiO_3^{2-} to the removal of Sb(V) had also been observed. Generally, the interference of these anions on the treatment performance followed the sorting as silicate > phosphate > carbonate.

3.5. Effect of flow rate in a continuous system

An appropriate selection of retention time is crucial to the running performance of the continuous process, which not only definitely controls the available time for the reactions between adsorbent and adsorbate, also directly affects the operating time and cost of the synergistic treatment. The effect of feed-water flow on the removal efficiencies of Sb(V) in the continuous tests is shown in Fig. 6. The effect of feed-water flow on the operating costs is listed in Table 4. A total of 6 L feed water with each concentration was pumped to the column device for each experiment. A horizontal axis at the removal efficiency of 90% was drawn to evaluate specific treatment capacity of the column process for each kind of Sb(V) concentration. As shown, the removal efficiencies in five groups all gradually decreased over the effluent volumes. The intersection of the horizontal axis at 90% and the curve of 'flow 40 mL/min' reached the maximum effluent volume (Table 4, i.e., 3.042 L) among the five points. This means the column of 'flow 40 mL/min' had cumulatively processed the maximum volume of feed water at the concentration of 50 mg/L when the removal efficiency of Sb(V) detected in the effluent water was just reduced to the level of 90%. As shown in Table 4, the corresponding minimum operating cost is 4,552.58 \$/m^3, obtaining still at the group of '40 mL/min'. Generally, the flow rate of 40 mL/min among the five rates facilitated the tandem units to achieve best removal performance in the continuous treatment process at a minimum operating cost.

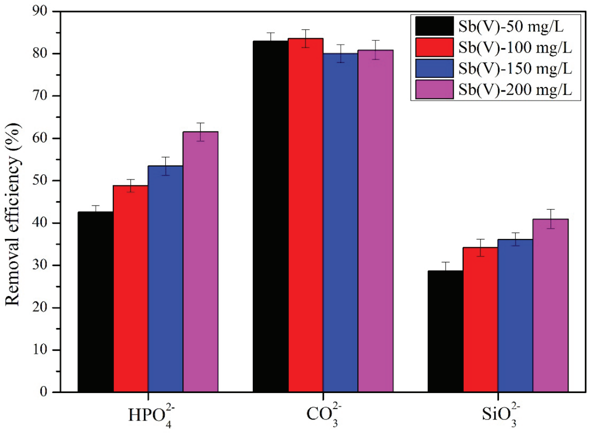

Fig. 5. Effect of competing ions on the removal efficiency of Sb(V) in the batch tests (Experimental conditions: at the voltage gradient of 1.0 V/cm, proposing time of 20 min, and initial pH of 4).

3.6. Mechanism discussion

Some stable Sb species are commonly formed through adsorption or precipitation on the oxyhydroxides in the natural systems. Some factors, such as pH, inorganic ligands and E_h, noticeably affect the sequestration or immobilization of Sb ions on the mineral surfaces. Similarly, the transformation of soluble Sb(V) to the insoluble species in the treatment process is also affected by these factors. The changes of pH and E_h over the proposing time (150 min) in the column-mode experiments are shown in Fig. 7. The results of two-tailed

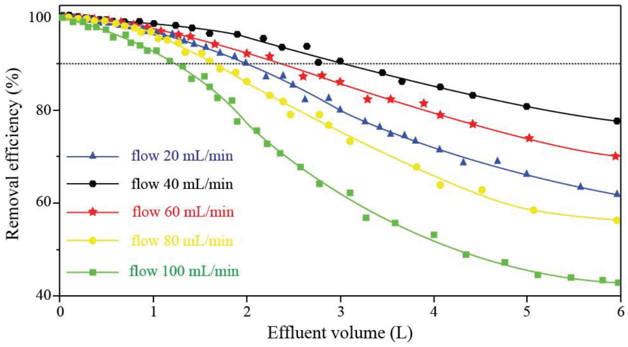

Fig. 6. Effect of feed-water flow on the removal efficiency of Sb(V) in the continuous tests (Experimental conditions: at the voltage gradient of 1.0 V/cm, initial pH of 4, and Sb(V) concentration of 50 mg/L).

Table 4
Effect of feed-water flow on the operating costs (at the voltage gradient of 1.0 V/cm, initial pH of 4 and Sb(V) concentration of 50 mg/L)

Flow rate (mL/min)	Treatment capacity at larger than $Re_{Sb(V)}$ of 90% (L)	Proposing time (min)	Operating costs (\$/ m^3)
20	2.025	101.25	6,839.45
40	3.042	76.05	4,552.58
60	2.375	39.58	5,830.42
80	1.656	20.7	8,361.55
100	1.251	12.51	11,068.25

t-tests for AC and AC-nZVI are shown in Table 5. The pH values flocculated around 4 for both adsorbents (Fig. 7(a)). Generally, the pH in the effluent was slightly elevated for AC-nZVI and marginally reduced for AC, compared with the initial pH value. The negatively charged Sb(V) species are expected in the presented pH ranges (i.e., pH > 2.7), no matter for AC or AC-nZVI filled process. However, pH_{zpc} changes (Fig. 2(a)) between the two kinds of adsorbents have utterly changed the adsorption of Sb(V) in the two systems. The electrostatic attraction and electrostatic repulsion had clearly dominated AC-nZVI and AC adsorption, respectively, which directly enlarges the equilibrium gap between the two systems (Fig. 2(b) and 2(c)). E_h values in AC-nZVI system highly increased from 382.2 to 598.8 mV after a moderate decrease from 424.8 to 382.2 mV at the initial 70 min. The change of E_h in AC system was comparatively smaller. The result means some nZVI particles had been dissolved from the carrier and further oxidized. Some soluble Sb(V) ions were sequestrated in the new formation of $FeSbO_4$ precipitation. Furthermore, during the EK process, the iron cations were electro-migrated to cathode and formed iron-based oxyhydroxides with the hydrolysis-generated OH- ions, which also immobilized some Sb(V) anions through co-precipitation. As shown, the significance probabilities (two-tailed) of pH and E_h between AC and AC-nZVI were 0.001 and 0.070, respectively, indicating the difference between the treatment systems was significant. The modification of AC remarkably enhanced Sb(V) removal from the stock solution. The effect of EK on the removal of Sb(V) was testified in the batch-mode experiments. The EK clearly strengthened the adsorption of Sb(V) based on the experimental results (Fig. 4.). To further elucidate the coupling effect of adsorption and EK on the removal of Sb(V), a treatment system without the adsorbents was constructed at a batch scale to monitor $Re_{Sb(V)}$ values under different voltage gradients. As shown in Fig. S2, $Re_{Sb(V)}$ values obtained in the AC-nZVI filling systems were obviously higher than the counterparts in the 'void' system at the same condition. The combination of EK and equilibrium adsorption undoubtedly maximized the removal of Sb(V) from feed water. The $Re_{Sb(V)}$ gradually increased with the elevations of voltage gradients in the system without any adsorbent, meaning the electromigration played an assignable role in influencing the solubilization behavior of Sb(V) during a dynamic process.

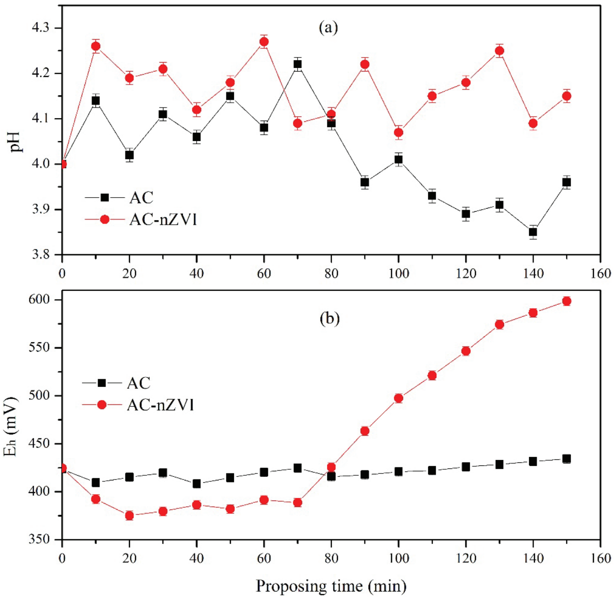

Fig. 7. Changes in pH (a) and E_h (b) over the proposing time in the continuous tests (Experimental conditions: at the voltage gradient of 1.0 V/cm, initial pH of 4, Sb(V) concentration of 50 mg/L, and feed-water flow of 40 mL/min).

3.7. Regeneration of adsorbent in the column

The regeneration capability of the column system was evaluated based on the removal experiments conducting in the column-mode tests with the reloading of the regenerated AC-nZVI. The regenerated AC-nZVI is continuously available for recycling use and reloads in the tests. The effect of regenerating cycles on the running performance of the column process is shown in Fig. 8. Clearly, the changing patterns of the top three curves including 'Raw AC-nZVI', 'First-cycle', and 'Second-cycle' were similar among the six

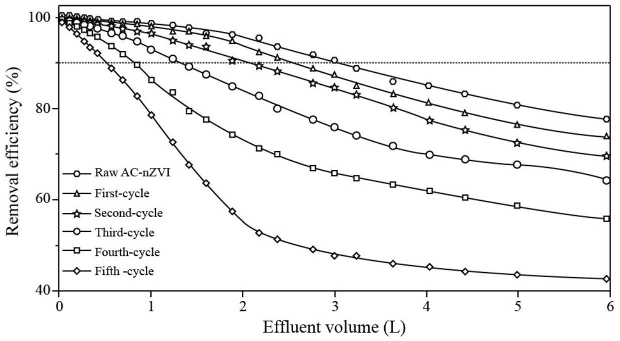

Fig. 8. Effect of regenerating cycles on the running performance of column process.

Table 5
Paired Samples-Tests for pH and E_h between AC and AC-nZVI

$A^a–B^b$	Mean	Paired differences				t	Df^c	Sig. (2-tailed)
		Std.deviation	Std. error mean	95% Confidence interval of the difference				
				Lower	Upper			
pH	–0.13500	0.12453	0.03113	–0.20136	–0.06864	–4.336	15	0.001
E_h	37.63125	77.07977	19.26994	–78.70416	3.44166	–1.953	15	0.070

^{a}AC, ^{b}AC-nZVI, ^{c}degrees of freedom.

curves. The removal efficiencies decreased more rapidly over the effluent volume with the increases in cycles. The intersections of the horizontal axis at the level of 90% $Re_{Sb(V)}$ and the removal curves were getting closer and closer to the ordinate axis. Comparably, the Sb(V) adsorption capacity dropped to 70% after the second cycle for La-doped magnetic biochar [26]. The Sb(V) removal rates still reached to 82% after 5 cycles for Fe-Mn binary oxides [27]. Generally, the reusability results indicated that the cycling increase gradually deteriorated the running performance of treatment units due to the diminishing of some active sites on the surfaces of adsorbents. However, the adsorbent being regenerated within three times is still acceptable and is considered to be qualified in reloading to the continuous treatment process in guaranteeing removal of Sb(V) species.

4. Conclusion

The combination of EK and equilibrium adsorption undoubtedly maximized Sb(V) removal from feed water. The feasibility of technology to removal Sb from the contaminated solution was fully evaluated in a continuous treatment. A sufficient but also a reasonable dose of AC-nZVI particles at pH of 4–5 was most conducive to achieve a maximal adsorption capacity (mg/g) of AC-nZVI in the equilibrium tests. The maximum monolayer-adsorption capacities of AC and AC-nZVI predicted by Langmuir isotherm were 10.26799 and 20.49180 $mg \cdot g^{-1}$, respectively. The changes in factors significantly affected removal efficiencies in a batch-mode treatment unit. In accordance with significance analysis and marginal means at a batch scale, the optimization combination of parameters was confirmed, namely 1.0 V/cm of voltage gradient, 20 min of proposing time, 4 of initial pH, and 50 mg/L of Sb(V) concentration, respectively. For the competing tests, the silicate and phosphate remarkably inhibited the adsorption of Sb(V) from feed water. The interference of competing anions on treatment performance for a coupling system followed a sorting as silicate > phosphate > carbonate. The choice of 40 mL/min flow rate was beneficial for the assembled units to reaching the best removal performance in a continuous treatment process at a minimum operating cost. Summarily, the mechanisms including the adsorption, precipitation, co-precipitation, and EK have comprehensively influenced the remediation of Sb(V) pollution in a continuous process. It is acceptable and qualified for the regenerated AC-nZVI being reloaded to the continuous treatment process within three recycling times. Obviously, the combination of the EK and iron-based adsorption is a method worth promoting in antimony removal and recycling from the wastewater. The feasibility analysis in a continuous process and the regeneration of adsorbent absolutely pave a way for an industrial-scale disposal.

Acknowledgement

The author received no financial support for the research, authorship, and/or publication of this article.

Conflict of interest

The co-authors have no conflict of interest to declare.

References

[1] H. Mubarak, L.Y. Chai, N. Mirza, Z.H. Yang, A. Pervez, M. Tariq, S. Shaheen, Q. Mahmood, Antimony (Sb) - pollution and removal techniques - critical assessment of technologies, Toxicol. Environ. Chem., 97 (2015) 1296–1318.

[2] Y.Y. Miao, F.C. Han, B.C. Pan, Y.J. Niu, G.Z. Nie, L. Lv, Antimony(V) removal from water by hydrated ferric oxides supported by calcite sand and polymeric anion exchanger, J. Environ. Sci.-China, 26 (2014) 307–314.

[3] M.A. Salam, R.M. Mohamed, Removal of antimony (III) by multi-walled carbon nanotubes from model solution and environmental samples, Chem. Eng. Res. Des., 91 (2013) 1352–1360.

[4] M. Perry, S. Wyllie, V. Prajapati, J. Menten, A. Raab, J. Feldmann, D. Chakraborti, S. Sundar, M. Boelaert, A. Picado, A. Fairlamb, Arsenic, antimony, and Leishmania: has arsenic contamination of drinking water in India led to treatment-resistant kala-azar?, Lancet, 385 (2015) 80–80.

[5] R. Cidu, R. Biddau, E. Dore, A. Vacca, L. Marini, Antimony in the soil-water-plant system at the Su Suergiu abandoned mine (Sardinia, Italy): strategies to mitigate contamination, Sci. Total Environ., 497 (2014) 319–331.

[6] E. Chmielewska, W. Tylus, M. Drabik, J. Majzlan, J. Kravcak, C. Williams, M. Caplovicova, L. Caplovic, Structure investigation of nano-FeO(OH) modified clinoptilolite tuff for antimony removal, Micropor. Mesopor. Mater., 248 (2017) 222–233.

[7] S. Ogawa, M. Katoh, T. Sato, Contribution of Hydroxyapatite and Ferrihydrite in Combined Applications for the Removal of Lead and Antimony from Aqueous Solutions, Water Air Soil Pollut., 225 (2014).

[8] X.Y. He, X.B. Min, X.B.A. Luo, Efficient Removal of Antimony (III, V) from Contaminated Water by Amino Modification of a Zirconium Metal-Organic Framework with Mechanism Study, J. Chem. Eng. Data, 62 (2017) 1519–1529.

[9] B.W. Ma, X. Wang, R.P. Liu, Z.L. Qi, W.A. Jefferson, H.C. Lan, H.J. Liu, J.H. Qu, Enhanced antimony(V) removal using synergistic effects of Fe hydrolytic flocs and ultrafiltration membrane with sludge discharge evaluation, Water Res., 121 (2017) 171–177.

[10] M. Katoh, K. Hashimoto, T. Sato, Lead and Antimony Removal from Contaminated Soil by Phytoremediation Combined with an Immobilization Material, Clean-Soil Air Water, 44 (2016) 1717–1724.

[11] C.Y. Xiong, T.H. Li, A.L. Dang, T.K. Zhao, H. Li, H.Q. Lv, Two-step approach of fabrication of three-dimensional MnO_2-graphene-carbon nanotube hybrid as a binder-free supercapacitor electrode, J. Power Sources, 306 (2016) 602–610.

[12] X. Du, F.S. Qu, H. Liang, K. Li, H.R. Yu, L.M. Bai, G.B. Li, Removal of antimony (III) from polluted surface water using a hybrid coagulation-flocculation-ultrafiltration (CF-UF) process, Chem. Eng. J., 254 (2014) 293–301.

[13] X.Z. Yang, T.Z. Zhou, B.Z. Ren, Z. Shi, A. Hursthouse, Synthesis, Characterization, and adsorptive properties of Fe_3O_4/GO nanocomposites for antimony removal, J. Anal. Methods Chem., 2017.

[14] J. Ilavsky, D. Barlokova, P. Hudec, K. Munka, Iron-based sorption materials for the removal of antimony from water, J. Water Supply Res. Technol. AQUA, 63 (2014) 518–524.

[15] B. Verbinnen, C. Block, P. Lievens, A. Van Brecht, C. Vandecasteele, Simultaneous removal of molybdenum, antimony and selenium oxyanions from wastewater by adsorption on supported magnetite, Waste Biomass Valorization, 4 (2013) 635–645.

[16] Z.L. Qi, T.P. Joshi, R.P. Liu, H.J. Liu, J.H. Qu, Synthesis of Ce(III)-doped Fe_3O_4 magnetic particles for efficient removal of antimony from aqueous solution, J. Hazard. Mater., 329 (2017) 193–204.

[17] J. Ilavsky, D. Barlokova, K. Munka, The Use of iron-based sorption materials and magnetic fields for the removal of antimony from water, Pol. J. Environ. Stud., 24 (2015) 1983–1992.

[18] T. Huang, L.F. Liu, L.L. Zhou, K. Yang, Operating optimization for the heavy metal removal from the municipal solid waste incineration fly ashes in the three-dimensional Chock tar electrokinetics, Chemosphere, 204 (2018) 294–302.

[19] T. Huang, L.F. Liu, L.L. Zhou, S.W. Zhang, Electrokinetic removal of chromium from chromite ore-processing residue using graphite particle-supported nanoscale zero-valent iron as the three-dimensional electrode, Chem. Eng. J., 350 (2018) 1022–1034.

[20] T. Huang, L.L. Zhou, L.F. Liu, M. Xia, Ultrasound-enhanced electrokinetic remediation for removal of Zn, Pb, Cu and Cd in municipal solid waste incineration fly ashes, Waste Manage., 75 (2018) 226–235.

[21] T. Huang, Q.K. Peng, L. Yu, D.W. Li, The Detoxification of heavy metals in the phosphate tailing-contaminated soil through sequential microbial pretreatment and electrokinetic remediation, Soil Sediment Contam., 26 (2017) 308–322.

[22] D.W. Li, T. Huang, K.X. Liu, Near-anode focusing phenomenon caused by the coupling effect of early precipitation and backward electromigration in electrokinetic remediation of MSWI fly ashes, Environ. Technol., 37 (2016) 216–227.

[23] T. Huang, D.W. Li, K.X. Liu, Y.W. Zhang, Heavy metal removal from MSWI fly ash by electrokinetic remediation coupled with a permeable activated charcoal reactive barrier, Sci. Rep., 5 (2015).

[24] K.X. Liu, T. Huang, X. Huang, L. Yu, F. Muhammad, B.Q. Jiao, D.W. Li, The application of homemade Neosinocalamus affinis AC in electrokinetic removal technology on heavy metal removal from the MSWI fly ash, Sci. Rep., 6 (2016).

[25] T. Huang, L.F. Liu, J.J. Tao, L.L. Zhou, S.W. Zhang, Microbial fuel cells coupling with the three-dimensional electro-Fenton technique enhances the degradation of methyl orange in the wastewater, Environ. Sci. Pollut. Res. Int., 25 (2018) 17989–18000.

[26] L. Wang, J.Y. Wang, Z.X. Wang, C. He, W. Lyu, W. Yan, L. Yang, Enhanced antimonate (Sb(V)) removal from aqueous solution by La-doped magnetic biochars, Chem. Eng. J., 354 (2018) 623–632.

[27] K.L. Yang, J.S. Zhou, Z.M. Lou, X.R. Zhou, Y.L. Liu, Y.Z. Li, S.A. Baig, X.H. Xu, Removal of Sb(V) from aqueous solutions using Fe-Mn binary oxides: the influence of iron oxides forms and the role of manganese oxides, Chem. Eng. J., 354 (2018) 577–588.

Supporting information

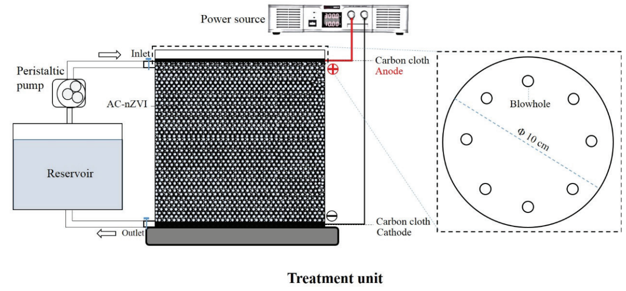

Fig. S1. Schematic cross-section of the cylindrical device.

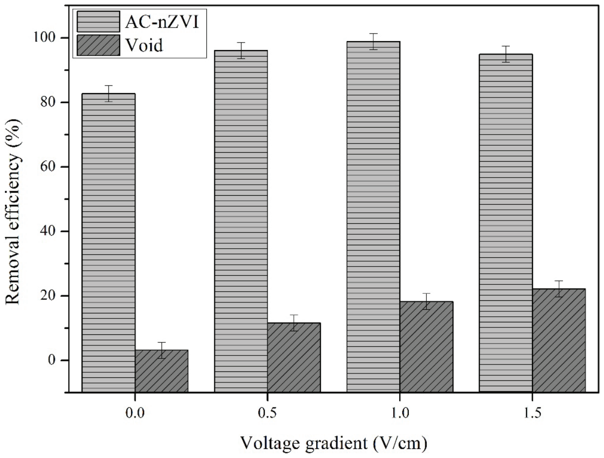

Fig. S2. Removal efficiencies of Sb(V) affected by the voltage gradients: AC-nZVI stacking system and void stacking system (Experimental conditions: at the initial pH of 4, Sb(V) concentration of 50 mg/L, proposing time of 20 min, and feed-water flow of 40 mL/min).

Table S1
Four factors at four different levels in the orthogonal tests

Level	A: Voltage gradient (V/cm)	B: Proposing time (min)	C: Initial pH	D: Sb(V) Concentration (mg/L)
1	0	5	4	50
2	0.5	10	6	100
3	1.0	15	8	150
4	1.5	20	10	200

Table S2
Estimated marginal means of Voltage gradient * Proposing time

Dependent variable: removal efficiency (%)

Voltage gradient (V/cm)	Proposing time (min)	Mean	Std. error	95% Confidence interval	
				Lower bound	Upper bound
0	5	73.112	0.709	70.857	75.368
	10	77.012	0.709	74.757	79.268
	15	78.687	0.709	76.432	80.943
	20	80.388	0.709	78.132	82.643
0.5	5	82.950	0.709	80.694	85.206
	10	86.850	0.709	84.594	89.106
	15	88.525	0.709	86.269	90.781
	20	90.225	0.709	87.969	92.481
1.0	5	87.620	0.709	85.364	89.876
	10	91.520	0.709	89.264	93.776
	15	93.195	0.709	90.939	95.451
	20	94.895	0.709	92.639	97.151
1.5	5	84.918	0.709	82.662	87.173
	10	88.818	0.709	86.562	91.073
	15	90.493	0.709	88.237	92.748
	20	92.193	0.709	89.937	94.448

Table S3
Estimated marginal means of Voltage gradient * Initial pH

Dependent variable: removal efficiency (%)					
Voltage gradient (V/cm)	Initial pH	Mean	Std. error	95% Confidence interval	
				Lower bound	Upper bound
0	4	78.932	0.709	76.677	81.188
	6	76.975	0.709	74.719	79.231
	8	78.490	0.709	76.234	80.746
	10	74.803	0.709	72.547	77.058
0.5	4	88.770	0.709	86.514	91.026
	6	86.812	0.709	84.557	89.068
	8	88.327	0.709	86.072	90.583
	10	84.640	0.709	82.384	86.896
1.0	4	93.440	0.709	91.184	95.696
	6	91.482	0.709	89.227	93.738
	8	92.997	0.709	90.742	95.253
	10	89.310	0.709	87.054	91.566
1.5	4	90.738	0.709	88.482	92.993
	6	88.780	0.709	86.524	91.036
	8	90.295	0.709	88.039	92.551
	10	86.608	0.709	84.352	88.863

Table S4
Estimated marginal means of Voltage gradient * Sb(V)-concentration

Dependent variable: removal efficiency (%)					
Voltage gradient (V/cm)	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
				Lower bound	Upper bound
0	50	81.330	0.709	79.074	83.586
	100	79.085	0.709	76.829	81.341
	150	75.367	0.709	73.112	77.623
	200	73.418	0.709	71.162	75.673
0.5	50	91.167	0.709	88.912	93.423
	100	88.922	0.709	86.667	91.178
	150	85.205	0.709	82.949	87.461
	200	83.255	0.709	80.999	85.511
1.0	50	95.837	0.709	93.582	98.093
	100	93.592	0.709	91.337	95.848
	150	89.875	0.709	87.619	92.131
	200	87.925	0.709	85.669	90.181
1.5	50	93.135	0.709	90.879	95.391
	100	90.890	0.709	88.634	93.146
	150	87.173	0.709	84.917	89.428
	200	85.223	0.709	82.967	87.478

Table S5
Estimated marginal means of Proposing time * Initial pH

Dependent variable: removal efficiency (%)

Proposing time (min)	Initial pH	Mean	Std. error	95% confidence interval	
				Lower bound	Upper bound
5	4	83.782	0.709	81.527	86.038
	6	81.825	0.709	79.569	84.081
	8	83.340	0.709	81.084	85.596
	10	79.653	0.709	77.397	81.908
10	4	87.682	0.709	85.427	89.938
	6	85.725	0.709	83.469	87.981
	8	87.240	0.709	84.984	89.496
	10	83.553	0.709	81.297	85.808
15	4	89.357	0.709	87.102	91.613
	6	87.400	0.709	85.144	89.656
	8	88.915	0.709	86.659	91.171
	10	85.228	0.709	82.972	87.483
20	4	91.058	0.709	88.802	93.313
	6	89.100	0.709	86.844	91.356
	8	90.615	0.709	88.359	92.871
	10	86.928	0.709	84.672	89.183

Table S6
Estimated marginal means of Proposing time * Sb(V)-concentration

Dependent variable: removal efficiency (%)

Proposing time (min)	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
				Lower bound	Upper bound
5	50	86.180	0.709	83.924	88.436
	100	83.935	0.709	81.679	86.191
	150	80.217	0.709	77.962	82.473
	200	78.268	0.709	76.012	80.523
10	50	90.080	0.709	87.824	92.336
	100	87.835	0.709	85.579	90.091
	150	84.117	0.709	81.862	86.373
	200	82.168	0.709	79.912	84.423
15	50	91.755	0.709	89.499	94.011
	100	89.510	0.709	87.254	91.766
	150	85.792	0.709	83.537	88.048
	200	83.843	0.709	81.587	86.098
20	50	93.455	0.709	91.199	95.711
	100	91.210	0.709	88.954	93.466
	150	87.493	0.709	85.237	89.748
	200	85.543	0.709	83.287	87.798

Table S7
Estimated marginal means of Initial pH * Sb(V)-concentration

Dependent variable: removal efficiency (%)

Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
				Lower bound	Upper bound
4	50	92.000	0.709	89.744	94.256
	100	89.755	0.709	87.499	92.011
	150	86.037	0.709	83.782	88.293
	200	84.088	0.709	81.832	86.343
6	50	90.042	0.709	87.787	92.298
	100	87.797	0.709	85.542	90.053
	150	84.080	0.709	81.824	86.336
	200	82.130	0.709	79.874	84.386
8	50	91.557	0.709	89.302	93.813
	100	89.312	0.709	87.057	91.568
	150	85.595	0.709	83.339	87.851
	200	83.645	0.709	81.389	85.901
10	50	87.870	0.709	85.614	90.126
	100	85.625	0.709	83.369	87.881
	150	81.908	0.709	79.652	84.163
	200	79.958	0.709	77.702	82.213

Table S8
Estimated marginal means of Voltage gradient * Proposing time * Initial pH

Dependent variable: removal efficiency (%)

Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
0	5	4	74.745	0.847	72.049	77.441
		6	72.787	0.847	70.091	75.484
		8	74.302	0.847	71.606	76.999
		10	70.615	0.847	67.919	73.311
	10	4	78.645	0.847	75.949	81.341
		6	76.687	0.847	73.991	79.384
		8	78.202	0.847	75.506	80.899
		10	74.515	0.847	71.819	77.211
	15	4	80.320	0.847	77.624	83.016
		6	78.362	0.847	75.666	81.059
		8	79.877	0.847	77.181	82.574
		10	76.190	0.847	73.494	78.886
	20	4	82.020	0.847	79.324	84.716
		6	80.063	0.847	77.366	82.759
		8	81.578	0.847	78.881	84.274
		10	77.890	0.847	75.194	80.586
0.5	5	4	84.582	0.847	81.886	87.279
		6	82.625	0.847	79.929	85.321
		8	84.140	0.847	81.444	86.836
		10	80.453	0.847	77.756	83.149
	10	4	88.482	0.847	85.786	91.179
		6	86.525	0.847	83.829	89.221
		8	88.040	0.847	85.344	90.736

(*Continued*)

Table S8 (*Continued*)

Dependent variable: removal efficiency (%)						
Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
		10	84.353	0.847	81.656	87.049
	15	4	90.157	0.847	87.461	92.854
		6	88.200	0.847	85.504	90.896
		8	89.715	0.847	87.019	92.411
		10	86.028	0.847	83.331	88.724
	20	4	91.858	0.847	89.161	94.554
		6	89.900	0.847	87.204	92.596
		8	91.415	0.847	88.719	94.111
		10	87.728	0.847	85.031	90.424
1.0	5	4	89.252	0.847	86.556	91.949
		6	87.295	0.847	84.599	89.991
		8	88.810	0.847	86.114	91.506
		10	85.123	0.847	82.426	87.819
	10	4	93.152	0.847	90.456	95.849
		6	91.195	0.847	88.499	93.891
		8	92.710	0.847	90.014	95.406
		10	89.023	0.847	86.326	91.719
	15	4	94.827	0.847	92.131	97.524
		6	92.870	0.847	90.174	95.566
		8	94.385	0.847	91.689	97.081
		10	90.698	0.847	88.001	93.394
	20	4	96.528	0.847	93.831	99.224
		6	94.570	0.847	91.874	97.266
		8	96.085	0.847	93.389	98.781
		10	92.398	0.847	89.701	95.094
1.5	5	4	86.550	0.847	83.854	89.246
		6	84.593	0.847	81.896	87.289
		8	86.108	0.847	83.411	88.804
		10	82.420	0.847	79.724	85.116
	10	4	90.450	0.847	87.754	93.146
		6	88.493	0.847	85.796	91.189
		8	90.008	0.847	87.311	92.704
		10	86.320	0.847	83.624	89.016
	15	4	92.125	0.847	89.429	94.821
		6	90.168	0.847	87.471	92.864
		8	91.683	0.847	88.986	94.379
		10	87.995	0.847	85.299	90.691
	20	4	93.825	0.847	91.129	96.521
		6	91.868	0.847	89.171	94.564
		8	93.383	0.847	90.686	96.079
		10	89.695	0.847	86.999	92.391

Table S9
Estimated marginal means of Voltage gradient * Proposing time * Sb(V)-concentration

Dependent variable: removal efficiency (%)						
Voltage gradient (V/cm)	Proposing time (min)	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
0	5	50	77.142	0.847	74.446	79.839
		100	74.897	0.847	72.201	77.594
		150	71.180	0.847	68.484	73.876
		200	69.230	0.847	66.534	71.926
	10	50	81.042	0.847	78.346	83.739
		100	78.797	0.847	76.101	81.494
		150	75.080	0.847	72.384	77.776
		200	73.130	0.847	70.434	75.826
	15	50	82.717	0.847	80.021	85.414
		100	80.472	0.847	77.776	83.169
		150	76.755	0.847	74.059	79.451
		200	74.805	0.847	72.109	77.501
	20	50	84.418	0.847	81.721	87.114
		100	82.173	0.847	79.476	84.869
		150	78.455	0.847	75.759	81.151
		200	76.505	0.847	73.809	79.201
0.5	5	50	86.980	0.847	84.284	89.676
		100	84.735	0.847	82.039	87.431
		150	81.017	0.847	78.321	83.714
		200	79.068	0.847	76.371	81.764
	10	50	90.880	0.847	88.184	93.576
		100	88.635	0.847	85.939	91.331
		150	84.917	0.847	82.221	87.614
		200	82.968	0.847	80.271	85.664
	15	50	92.555	0.847	89.859	95.251
		100	90.310	0.847	87.614	93.006
		150	86.592	0.847	83.896	89.289
		200	84.643	0.847	81.946	87.339
	20	50	94.255	0.847	91.559	96.951
		100	92.010	0.847	89.314	94.706
		150	88.293	0.847	85.596	90.989
		200	86.343	0.847	83.646	89.039
1.0	5	50	91.650	0.847	88.954	94.346
		100	89.405	0.847	86.709	92.101
		150	85.687	0.847	82.991	88.384
		200	83.738	0.847	81.041	86.434
	10	50	95.550	0.847	92.854	98.246
		100	93.305	0.847	90.609	96.001
		150	89.587	0.847	86.891	92.284
		200	87.638	0.847	84.941	90.334
	15	50	97.225	0.847	94.529	99.921
		100	94.980	0.847	92.284	97.676
		150	91.262	0.847	88.566	93.959
		200	89.313	0.847	86.616	92.009
	20	50	98.925	0.847	96.229	101.621
		100	96.680	0.847	93.984	99.376

(*Continued*)

Table S9 (*Continued*)

Dependent variable: removal efficiency (%)

Voltage gradient (V/cm)	Proposing time (min)	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
		150	92.963	0.847	90.266	95.659
		200	91.013	0.847	88.316	93.709
1.5	5	50	88.948	0.847	86.251	91.644
		100	86.703	0.847	84.006	89.399
		150	82.985	0.847	80.289	85.681
		200	81.035	0.847	78.339	83.731
	10	50	92.848	0.847	90.151	95.544
		100	90.603	0.847	87.906	93.299
		150	86.885	0.847	84.189	89.581
		200	84.935	0.847	82.239	87.631
	15	50	94.523	0.847	91.826	97.219
		100	92.278	0.847	89.581	94.974
		150	88.560	0.847	85.864	91.256
		200	86.610	0.847	83.914	89.306
	20	50	96.223	0.847	93.526	98.919
		100	93.978	0.847	91.281	96.674
		150	90.260	0.847	87.564	92.956
		200	88.310	0.847	85.614	91.006

Table S10
Estimated marginal means of Voltage gradient * Initial pH * Sb(V)-concentration

Dependent variable: removal efficiency (%)

Voltage gradient (V/cm)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
0	4	50	82.962	0.847	80.266	85.659
		100	80.717	0.847	78.021	83.414
		150	77.000	0.847	74.304	79.696
		200	75.050	0.847	72.354	77.746
	6	50	81.005	0.847	78.309	83.701
		100	78.760	0.847	76.064	81.456
		150	75.042	0.847	72.346	77.739
		200	73.093	0.847	70.396	75.789
	8	50	82.520	0.847	79.824	85.216
		100	80.275	0.847	77.579	82.971
		150	76.557	0.847	73.861	79.254
		200	74.608	0.847	71.911	77.304
	10	50	78.833	0.847	76.136	81.529
		100	76.588	0.847	73.891	79.284
		150	72.870	0.847	70.174	75.566
		200	70.920	0.847	68.224	73.616
0.5	4	50	92.800	0.847	90.104	95.496
		100	90.555	0.847	87.859	93.251
		150	86.837	0.847	84.141	89.534
		200	84.888	0.847	82.191	87.584
	6	50	90.842	0.847	88.146	93.539
		100	88.597	0.847	85.901	91.294

(*Continued*)

Table S10 (*Continued*)

Dependent variable: removal efficiency (%)						
Voltage gradient (V/cm)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
		150	84.880	0.847	82.184	87.576
		200	82.930	0.847	80.234	85.626
	8	50	92.357	0.847	89.661	95.054
		100	90.112	0.847	87.416	92.809
		150	86.395	0.847	83.699	89.091
		200	84.445	0.847	81.749	87.141
	10	50	88.670	0.847	85.974	91.366
		100	86.425	0.847	83.729	89.121
		150	82.708	0.847	80.011	85.404
		200	80.758	0.847	78.061	83.454
1.0	4	50	97.470	0.847	94.774	100.166
		100	95.225	0.847	92.529	97.921
		150	91.507	0.847	88.811	94.204
		200	89.558	0.847	86.861	92.254
	6	50	95.512	0.847	92.816	98.209
		100	93.267	0.847	90.571	95.964
		150	89.550	0.847	86.854	92.246
		200	87.600	0.847	84.904	90.296
	8	50	97.027	0.847	94.331	99.724
		100	94.782	0.847	92.086	97.479
		150	91.065	0.847	88.369	93.761
		200	89.115	0.847	86.419	91.811
	10	50	93.340	0.847	90.644	96.036
		100	91.095	0.847	88.399	93.791
		150	87.378	0.847	84.681	90.074
		200	85.428	0.847	82.731	88.124
1.5	4	50	94.768	0.847	92.071	97.464
		100	92.523	0.847	89.826	95.219
		150	88.805	0.847	86.109	91.501
		200	86.855	0.847	84.159	89.551
	6	50	92.810	0.847	90.114	95.506
		100	90.565	0.847	87.869	93.261
		150	86.848	0.847	84.151	89.544
		200	84.898	0.847	82.201	87.594
	8	50	94.325	0.847	91.629	97.021
		100	92.080	0.847	89.384	94.776
		150	88.363	0.847	85.666	91.059
		200	86.413	0.847	83.716	89.109
	10	50	90.638	0.847	87.941	93.334
		100	88.393	0.847	85.696	91.089
		150	84.675	0.847	81.979	87.371
		200	82.725	0.847	80.029	85.421

Table S11
Estimated marginal means of Proposing time * Initial pH * Sb(V)-concentration

Dependent variable: removal efficiency (%)

Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
5	4	50	87.812	0.847	85.116	90.509
		100	85.567	0.847	82.871	88.264
		150	81.850	0.847	79.154	84.546
		200	79.900	0.847	77.204	82.596
	6	50	85.855	0.847	83.159	88.551
		100	83.610	0.847	80.914	86.306
		150	79.892	0.847	77.196	82.589
		200	77.943	0.847	75.246	80.639
	8	50	87.370	0.847	84.674	90.066
		100	85.125	0.847	82.429	87.821
		150	81.407	0.847	78.711	84.104
		200	79.458	0.847	76.761	82.154
	10	50	83.683	0.847	80.986	86.379
		100	81.438	0.847	78.741	84.134
		150	77.720	0.847	75.024	80.416
		200	75.770	0.847	73.074	78.466
10	4	50	91.712	0.847	89.016	94.409
		100	89.467	0.847	86.771	92.164
		150	85.750	0.847	83.054	88.446
		200	83.800	0.847	81.104	86.496
	6	50	89.755	0.847	87.059	92.451
		100	87.510	0.847	84.814	90.206
		150	83.792	0.847	81.096	86.489
		200	81.843	0.847	79.146	84.539
	8	50	91.270	0.847	88.574	93.966
		100	89.025	0.847	86.329	91.721
		150	85.307	0.847	82.611	88.004
		200	83.358	0.847	80.661	86.054
	10	50	87.583	0.847	84.886	90.279
		100	85.338	0.847	82.641	88.034
		150	81.620	0.847	78.924	84.316
		200	79.670	0.847	76.974	82.366
15	4	50	93.387	0.847	90.691	96.084
		100	91.142	0.847	88.446	93.839
		150	87.425	0.847	84.729	90.121
		200	85.475	0.847	82.779	88.171
	6	50	91.430	0.847	88.734	94.126
		100	89.185	0.847	86.489	91.881
		150	85.467	0.847	82.771	88.164
		200	83.518	0.847	80.821	86.214
	8	50	92.945	0.847	90.249	95.641
		100	90.700	0.847	88.004	93.396
		150	86.982	0.847	84.286	89.679
		200	85.033	0.847	82.336	87.729
	10	50	89.258	0.847	86.561	91.954
		100	87.013	0.847	84.316	89.709

(*Continued*)

Table S11 (*Continued*)

Dependent variable: removal efficiency (%)						
Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
					Lower bound	Upper bound
		150	83.295	0.847	80.599	85.991
		200	81.345	0.847	78.649	84.041
20	4	50	95.088	0.847	92.391	97.784
		100	92.843	0.847	90.146	95.539
		150	89.125	0.847	86.429	91.821
		200	87.175	0.847	84.479	89.871
	6	50	93.130	0.847	90.434	95.826
		100	90.885	0.847	88.189	93.581
		150	87.168	0.847	84.471	89.864
		200	85.218	0.847	82.521	87.914
	8	50	94.645	0.847	91.949	97.341
		100	92.400	0.847	89.704	95.096
		150	88.683	0.847	85.986	91.379
		200	86.733	0.847	84.036	89.429
	10	50	90.958	0.847	88.261	93.654
		100	88.713	0.847	86.016	91.409
		150	84.995	0.847	82.299	87.691
		200	83.045	0.847	80.349	85.741

Table S12

Estimated marginal means of Voltage gradient * Proposing time * Initial pH * Sb(V)-concentration

Dependent variable: removal efficiency (%)							
Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
						Lower bound	Upper bound
0	5	4	50	78.775	0.966	75.701	81.849
			100	76.530	0.966	73.456	79.604
			150	72.812	0.966	69.738	75.887
			200	70.863	0.966	67.788	73.937
		6	50	76.817	0.966	73.743	79.892
			100	74.572	0.966	71.498	77.647
			150	70.855	0.966	67.781	73.929
			200	68.905	0.966	65.831	71.979
		8	50	78.332	0.966	75.258	81.407
			100	76.087	0.966	73.013	79.162
			150	72.370	0.966	69.296	75.444
			200	70.420	0.966	67.346	73.494
		10	50	74.645	0.966	71.571	77.719
			100	72.400	0.966	69.326	75.474
			150	68.683	0.966	65.608	71.757
			200	66.733	0.966	63.658	69.807
	10	4	50	82.675	0.966	79.601	85.749
			100	80.430	0.966	77.356	83.504
			150	76.712	0.966	73.638	79.787

(*Continued*)

Table S12 (*Continued*)

Dependent variable: removal efficiency (%)

Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
						Lower bound	Upper bound
			200	74.763	0.966	71.688	77.837
		6	50	80.717	0.966	77.643	83.792
			100	78.472	0.966	75.398	81.547
			150	74.755	0.966	71.681	77.829
			200	72.805	0.966	69.731	75.879
		8	50	82.232	0.966	79.158	85.307
			100	79.987	0.966	76.913	83.062
			150	76.270	0.966	73.196	79.344
			200	74.320	0.966	71.246	77.394
		10	50	78.545	0.966	75.471	81.619
			100	76.300	0.966	73.226	79.374
			150	72.583	0.966	69.508	75.657
			200	70.633	0.966	67.558	73.707
	15	4	50	84.350	0.966	81.276	87.424
			100	82.105	0.966	79.031	85.179
			150	78.387	0.966	75.313	81.462
			200	76.438	0.966	73.363	79.512
		6	50	82.392	0.966	79.318	85.467
			100	80.147	0.966	77.073	83.222
			150	76.430	0.966	73.356	79.504
			200	74.480	0.966	71.406	77.554
		8	50	83.907	0.966	80.833	86.982
			100	81.662	0.966	78.588	84.737
			150	77.945	0.966	74.871	81.019
			200	75.995	0.966	72.921	79.069
		10	50	80.220	0.966	77.146	83.294
			100	77.975	0.966	74.901	81.049
			150	74.258	0.966	71.183	77.332
			200	72.308	0.966	69.233	75.382
	20	4	50	86.050	0.966	82.976	89.124
			100	83.805	0.966	80.731	86.879
			150	80.088	0.966	77.013	83.162
			200	78.138	0.966	75.063	81.212
		6	50	84.093	0.966	81.018	87.167
			100	81.848	0.966	78.773	84.922
			150	78.130	0.966	75.056	81.204
			200	76.180	0.966	73.106	79.254
		8	50	85.608	0.966	82.533	88.682
			100	83.363	0.966	80.288	86.437
			150	79.645	0.966	76.571	82.719
			200	77.695	0.966	74.621	80.769
		10	50	81.920	0.966	78.846	84.994
			100	79.675	0.966	76.601	82.749
			150	75.958	0.966	72.883	79.032
			200	74.008	0.966	70.933	77.082

(*Continued*)

Table S12 (*Continued*)

Dependent variable: removal efficiency (%)

Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
						Lower bound	Upper bound
0.5	5	4	50	88.612	0.966	85.538	91.687
			100	86.367	0.966	83.293	89.442
			150	82.650	0.966	79.576	85.724
			200	80.700	0.966	77.626	83.774
		6	50	86.655	0.966	83.581	89.729
			100	84.410	0.966	81.336	87.484
			150	80.692	0.966	77.618	83.767
			200	78.743	0.966	75.668	81.817
		8	50	88.170	0.966	85.096	91.244
			100	85.925	0.966	82.851	88.999
			150	82.207	0.966	79.133	85.282
			200	80.258	0.966	77.183	83.332
		10	50	84.483	0.966	81.408	87.557
			100	82.238	0.966	79.163	85.312
			150	78.520	0.966	75.446	81.594
			200	76.570	0.966	73.496	79.644
	10	4	50	92.512	0.966	89.438	95.587
			100	90.267	0.966	87.193	93.342
			150	86.550	0.966	83.476	89.624
			200	84.600	0.966	81.526	87.674
		6	50	90.555	0.966	87.481	93.629
			100	88.310	0.966	85.236	91.384
			150	84.592	0.966	81.518	87.667
			200	82.643	0.966	79.568	85.717
		8	50	92.070	0.966	88.996	95.144
			100	89.825	0.966	86.751	92.899
			150	86.107	0.966	83.033	89.182
			200	84.158	0.966	81.083	87.232
		10	50	88.383	0.966	85.308	91.457
			100	86.138	0.966	83.063	89.212
			150	82.420	0.966	79.346	85.494
			200	80.470	0.966	77.396	83.544
	15	4	50	94.187	0.966	91.113	97.262
			100	91.942	0.966	88.868	95.017
			150	88.225	0.966	85.151	91.299
			200	86.275	0.966	83.201	89.349
		6	50	92.230	0.966	89.156	95.304
			100	89.985	0.966	86.911	93.059
			150	86.267	0.966	83.193	89.342
			200	84.318	0.966	81.243	87.392
		8	50	93.745	0.966	90.671	96.819
			100	91.500	0.966	88.426	94.574
			150	87.782	0.966	84.708	90.857
			200	85.833	0.966	82.758	88.907
		10	50	90.058	0.966	86.983	93.132
			100	87.813	0.966	84.738	90.887
			150	84.095	0.966	81.021	87.169
			200	82.145	0.966	79.071	85.219

(*Continued*)

Table S12 (*Continued*)

Dependent variable: removal efficiency (%)							
Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
						Lower bound	Upper bound
	20	4	50	95.888	0.966	92.813	98.962
			100	93.643	0.966	90.568	96.717
			150	89.925	0.966	86.851	92.999
			200	87.975	0.966	84.901	91.049
		6	50	93.930	0.966	90.856	97.004
			100	91.685	0.966	88.611	94.759
			150	87.968	0.966	84.893	91.042
			200	86.018	0.966	82.943	89.092
		8	50	95.445	0.966	92.371	98.519
			100	93.200	0.966	90.126	96.274
			150	89.483	0.966	86.408	92.557
			200	87.533	0.966	84.458	90.607
		10	50	91.758	0.966	88.683	94.832
			100	89.513	0.966	86.438	92.587
			150	85.795	0.966	82.721	88.869
			200	83.845	0.966	80.771	86.919
1.0	5	4	50	93.282	0.966	90.208	96.357
			100	91.037	0.966	87.963	94.112
			150	87.320	0.966	84.246	90.394
			200	85.370	0.966	82.296	88.444
		6	50	91.325	0.966	88.251	94.399
			100	89.080	0.966	86.006	92.154
			150	85.362	0.966	82.288	88.437
			200	83.413	0.966	80.338	86.487
		8	50	92.840	0.966	89.766	95.914
			100	90.595	0.966	87.521	93.669
			150	86.877	0.966	83.803	89.952
			200	84.928	0.966	81.853	88.002
		10	50	89.153	0.966	86.078	92.227
			100	86.908	0.966	83.833	89.982
			150	83.190	0.966	80.116	86.264
			200	81.240	0.966	78.166	84.314
	10	4	50	97.182	0.966	94.108	100.257
			100	94.937	0.966	91.863	98.012
			150	91.220	0.966	88.146	94.294
			200	89.270	0.966	86.196	92.344
		6	50	95.225	0.966	92.151	98.299
			100	92.980	0.966	89.906	96.054
			150	89.262	0.966	86.188	92.337
			200	87.313	0.966	84.238	90.387
		8	50	96.740	0.966	93.666	99.814
			100	94.495	0.966	91.421	97.569
			150	90.777	0.966	87.703	93.852
			200	88.828	0.966	85.753	91.902
		10	50	93.053	0.966	89.978	96.127
			100	90.808	0.966	87.733	93.882
			150	87.090	0.966	84.016	90.164
			200	85.140	0.966	82.066	88.214

(*Continued*)

Table S12 (*Continued*)

Dependent variable: removal efficiency (%)

Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
						Lower bound	Upper bound
	15	4	50	98.857	0.966	95.783	101.932
			100	96.612	0.966	93.538	99.687
			150	92.895	0.966	89.821	95.969
			200	90.945	0.966	87.871	94.019
		6	50	96.900	0.966	93.826	99.974
			100	94.655	0.966	91.581	97.729
			150	90.937	0.966	87.863	94.012
			200	88.988	0.966	85.913	92.062
		8	50	98.415	0.966	95.341	101.489
			100	96.170	0.966	93.096	99.244
			150	92.452	0.966	89.378	95.527
			200	90.503	0.966	87.428	93.577
		10	50	94.728	0.966	91.653	97.802
			100	92.483	0.966	89.408	95.557
			150	88.765	0.966	85.691	91.839
			200	86.815	0.966	83.741	89.889
	20	4	50	100.558	0.966	97.483	103.632
			100	98.313	0.966	95.238	101.387
			150	94.595	0.966	91.521	97.669
			200	92.645	0.966	89.571	95.719
		6	50	98.600	0.966	95.526	101.674
			100	96.355	0.966	93.281	99.429
			150	92.638	0.966	89.563	95.712
			200	90.688	0.966	87.613	93.762
		8	50	100.115	0.966	97.041	103.189
			100	97.870	0.966	94.796	100.944
			150	94.153	0.966	91.078	97.227
			200	92.203	0.966	89.128	95.277
		10	50	96.428	0.966	93.353	99.502
			100	94.183	0.966	91.108	97.257
			150	90.465	0.966	87.391	93.539
			200	88.515	0.966	85.441	91.589
1.5	5	4	50	90.580	0.966	87.506	93.654
			100	88.335	0.966	85.261	91.409
			150	84.618	0.966	81.543	87.692
			200	82.668	0.966	79.593	85.742
		6	50	88.623	0.966	85.548	91.697
			100	86.378	0.966	83.303	89.452
			150	82.660	0.966	79.586	85.734
			200	80.710	0.966	77.636	83.784
		8	50	90.138	0.966	87.063	93.212
			100	87.893	0.966	84.818	90.967
			150	84.175	0.966	81.101	87.249
			200	82.225	0.966	79.151	85.299
		10	50	86.450	0.966	83.376	89.524
			100	84.205	0.966	81.131	87.279
			150	80.488	0.966	77.413	83.562
			200	78.538	0.966	75.463	81.612

(*Continued*)

Table S12 (*Continued*)

Dependent variable: removal efficiency (%)							
Voltage gradient (V/cm)	Proposing time (min)	Initial pH	Sb(V) Concentration (mg/L)	Mean	Std. error	95% Confidence interval	
						Lower bound	Upper bound
	10	4	50	94.480	0.966	91.406	97.554
			100	92.235	0.966	89.161	95.309
			150	88.518	0.966	85.443	91.592
			200	86.568	0.966	83.493	89.642
		6	50	92.523	0.966	89.448	95.597
			100	90.278	0.966	87.203	93.352
			150	86.560	0.966	83.486	89.634
			200	84.610	0.966	81.536	87.684
		8	50	94.038	0.966	90.963	97.112
			100	91.793	0.966	88.718	94.867
			150	88.075	0.966	85.001	91.149
			200	86.125	0.966	83.051	89.199
		10	50	90.350	0.966	87.276	93.424
			100	88.105	0.966	85.031	91.179
			150	84.388	0.966	81.313	87.462
			200	82.438	0.966	79.363	85.512
	15	4	50	96.155	0.966	93.081	99.229
			100	93.910	0.966	90.836	96.984
			150	90.193	0.966	87.118	93.267
			200	88.243	0.966	85.168	91.317
		6	50	94.198	0.966	91.123	97.272
			100	91.953	0.966	88.878	95.027
			150	88.235	0.966	85.161	91.309
			200	86.285	0.966	83.211	89.359
		8	50	95.713	0.966	92.638	98.787
			100	93.468	0.966	90.393	96.542
			150	89.750	0.966	86.676	92.824
			200	87.800	0.966	84.726	90.874
		10	50	92.025	0.966	88.951	95.099
			100	89.780	0.966	86.706	92.854
			150	86.063	0.966	82.988	89.137
			200	84.113	0.966	81.038	87.187
	20	4	50	97.855	0.966	94.781	100.929
			100	95.610	0.966	92.536	98.684
			150	91.893	0.966	88.818	94.967
			200	89.943	0.966	86.868	93.017
		6	50	95.898	0.966	92.823	98.972
			100	93.653	0.966	90.578	96.727
			150	89.935	0.966	86.861	93.009
			200	87.985	0.966	84.911	91.059
		8	50	97.413	0.966	94.338	100.487
			100	95.168	0.966	92.093	98.242
			150	91.450	0.966	88.376	94.524
			200	89.500	0.966	86.426	92.574
		10	50	93.725	0.966	90.651	96.799
			100	91.480	0.966	88.406	94.554
			150	87.763	0.966	84.688	90.837
			200	85.813	0.966	82.738	88.887

Isothermal models

The models including Langmuir (Eq. (S1)) and Ducbinin–Radushkevich (D-R, Eq. (S2)) were adopted to evaluate Q_m (mg/g) of adsorbent, where C_e is the equilibrium concentration (mg/L) of Sb(V), q_e (Eq. (S3)) is the equilibrium adsorption capacity (mg/g), q_0 is the maximum monolayer adsorption capacity (mg·g^{-1}), and q_m represents the theoretical saturation capacity (mg·g^{-1}), $\varepsilon = \mathrm{RTln}(1+1/C_e)$, T is the absolute temperature (K) and R is the universal gas constant (8.314 J·mol^{-1}·K^{-1}).

$$\frac{c_e}{q_e} = \frac{1}{q_0 k_l} + \frac{c_e}{q_0} \tag{S1}$$

$$\ln\left(q_e\right) = \ln\left(q_m\right) - \mathrm{B}\varepsilon^2 \tag{S2}$$

$$q_e = \frac{\left(c_0 - c_e\right)V}{m} \tag{S3}$$