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A dual-functional electroactive filter towards simultaneously Sb(III) oxidation and sequestration

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Dear Prof. Wu,

We greatly appreciate the editorial comments on our manuscript entitled "*A dual-functional electroactive filter towards simultaneously Sb(III) oxidation and sequestration*".

The editorial comments and suggestions have been taken into account in our manuscript revision. In this response letter, we first itemized the comments/suggestions raised by the editor, followed by our response and descriptions of the changes made to the original manuscript.

We hope that the revision is satisfactory to all.

Sincerely,

Yanbiao Liu

Professor

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TITLE:

A Dual-Functional Electroactive Filter Towards Simultaneously Sb(III) Oxidation and Sequestration

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KEYWORDS:

Sb(III), electroactive filter, dual-functional, titanate nanowires, carbon nanotubes, adsorption

SHORT ABSTRACT:

A protocol for the rational design of a dual-functional electroactive filter consisting of carbon nanotubes and titanate nanowires is reported and their environmental applications towards Sb(III) oxidation and sequestration is presented.

LONG ABSTRACT:

We have designed a facile method to synthesize a dual-functional electrochemical filter consisting of two 1-D materials: titanate nanowires and carbon nanotubes. The hybrid titanate-CNT filter was prepared by a sonication coupled with a post-filtration route. Due to the synergistic effects of the increased number of exposed sorption sites, electrochemical reactivity, small pore size of the titanate-CNT network coupled with a flow-through design, simultaneous Sb(III) oxidation and sequestration can be readily achieved. Atomic fluorescence spectrometer technology demonstrated that the applied electrical field accelerates the Sb(III) conversion rate and the as-obtained Sb(V) were adsorbed effectively by the titanate nanowires due to their Sb specificity. This protocol provides a practical solution for the removal of highly toxic Sb(III) and other similar heavy metal ions.

INTRODUCTION:

Recently, the environmental pollution caused by emerging antimony (Sb) has attracted much attention[1,2]. Extensive studies demonstrate that Sb compounds pose high toxicity to human and microorganisms, although present in low concentrations in the environment[3,4]. Even worse, conventional physicochemical or biological methods are usually ineffective to remove these emerging contaminants due to their low concentrations and high toxicity[5]. The most abundant species of Sb are Sb(V) and Sb(III), of which the latter form is more toxic.

Among the currently available treatment methods, adsorption is believed to be a promising and feasible alternative due to its high efficiency, low cost, and simplicity[6,7]. Till now, several nanoscale sorbents with tunable microstructures, large specific surface area and Sb specificity have been developed, such as TiO_2[8], MnO_2[9], titanate[10], zerovalent iron[11], iron oxides and other binary metal oxides[12,13]. A common problem when dealing with nanoscale adsorbents is the post-separation issue due to their small particle size. One strategy to address this issue is to load these nano-sorbents onto macro/micro-scale supports[14]. Another challenging issue restricting the wide application of adsorption technology is the poor mass transport caused by limited concentration of target compounds/molecules[15]. This issue may be partially addressed by adopting a membrane design and convention could enhance the mass transport significantly. Recent efforts have been devoted to develop advanced treatment systems that combine adsorption and oxidation in a single unit for effective Sb(III) removal. Here, we show how an electroactive titanate-carbon nanotube (titanate-CNT) filter was rationally designed and applied for the simultaneously adsorption and sequestration of toxic Sb(III). By fine-tuning the titanate loading amount, applied voltage, and flow rate, we demonstrate how the Sb(III) oxidation rate and sequestration efficiency can be tailored correspondingly. Although the fabrication and application of the electroactive filter is shown in this protocol, similar designs can also apply to the treatment of other heavy metal ions.

Minor changes in the fabrication process and reagents may cause significant changes in the morphology and performance of the final system. For instance, the hydrothermal time, temperature, and chemical purity have been shown to affect the microstructures of these nanoscale adsorbents. The flow rate of the adsorbate solution also determines the residence time within a flow-through system as well as the removal efficiency of target compounds. With clear identification of these key impacting parameters, a reproducible synthesis protocol can be secured and a stable removal efficiency of Sb(III) can be achieved. This protocol aims to provide detailed experience on the fabrication of dual-functional hybrid filters as well as their applications towards the removal of toxic heavy metal ions in a flow-through manner.

PROTOCOL:

CAUTION: Please carefully read relevant safety data sheets (SDS) of all chemicals and wear proper personal protection equipment (PPE) before use. Some of the chemicals are toxic and irritant. Be careful when handling carbon nanotubes, which may have additional hazards if inhaled or contacted by skin.

1. Preparation of the electroactive titanate-CNT filter

1.1. Preparation of titanate nanowires[16]

1.1.1. Dissolve 56 g of potassium hydroxide (KOH) in 100 mL of deionized water under vigorous stirring.

1.1.2. Add 3 g of titanium dioxide (TiO_2) powder into the as-dissolved KOH solution.

1.1.3. Transfer the above solution into a Teflon-lined reactor and keep it at 200 °C for 24 h.

1.1.4. Wash the obtained white precipitate with 0.1 mol/L hydrochloric acid (HCl) and deionized water until a neutral effluent pH is obtained. Dry the product under vacuum at 60 °C overnight.

1.1.5. Transfer the products to a tube furnace and heat it to 600 °C for 2 h with a ramp rate of 1 °C/min.

1.2. Preparation of titanate-CNT filter[17]

1.2.1. Add 20 mg of carbon nanotubes (CNTs) into 40 mL of n-methyl pyrrolidone (NMP). Probe-sonication for 40 min to obtain homogeneous solution.

1.2.2. Separately, add 20 mg of the as made titanate nanowires into 20 mL of NMP. Perform probe-sonication for 20 min.

1.2.3. Mix the titanate dispersion solution with the CNT dispersion solution. Filter the mixture solution onto a PTFE membrane, which serves as a support for the titanate-CNT filter.

1.2.4. Rinse sequentially with 100 mL of ethanol and 200 mL of deionized water.

NOTE: A CNT-alone filter can be prepared by a similar route without the addition of titanate nanowires.

2. Electrochemical filtration of Sb(III)

2.1. Description on the experimental apparatus[18]

2.1.1. Conduct the sorption experiments in an electrochemistry modified polycarbonate filtration casing (see **Figure 1**).

2.1.2. Use a DC power supply to drive the electrochemistry.

2.1.3. Adopt perforated titanium ring as connector for anodic or cathodic filters.

2.1.4. Use an insulating silicone rubber as a separator and seal.

2.2. Filtration experiments

2.2.1. Add 2.2 mg of $C_8H_4K_2O_{12}Sb_2.3H_2O$ into 1000 mL of deionized water to prepare 800 µg/L Sb(III) solution.

2.2.2. Transfer 100 mL of Sb(III) solution to a 150 mL beaker. Adjust solution pH to 7.

2.2.3. Place the as-prepared titanate-CNT filter anode into the polycarbonate filtration casing and place another CNT-alone filter as cathode. Seal the casing.

2.2.4. Pass through the filtration system with Sb(III) solution at a given flow. Apply a DC voltage during filtration.

2.2.5. Determine the Sb_{total} and Sb(III) concentration with atomic fluorescence spectrometer technique[17].

NOTE: In this process, flow rate and applied voltage can be tuned by a peristaltic pump and a DC power supply, respectively.

REPRESENTATIVE RESULTS:

The electroactive filtration apparatus employed is an electrochemically modified polycarbonate filtration casing (**Figure 1**). Field emission scanning electron microscope (FESEM) and transmission electron microscopy (TEM) techniques are employed to characterize the morphology of the titanate-CNT filter (**Figure 2**). To demonstrate the efficacy of the electrochemical filtration system, the change of Sb_{total} and Sb valence state as a function of time is determined (**Figure 3**).

The FESEM images of titanate-CNT filter suggest a roughened surface. TEM characterization suggests that these CNTs are entangled with titanate nanowires. This suggests that we have successfully synthesized the titanate-CNT hybrid materials (**Figure 2**).

The change of Sb_{total} and Sb valence state as a function of time at 2 V are examined (**Figure 3**). Results suggest that the Sb(V) concentration rises sharply within the initial 0.5 h and complete Sb(III) conversion is observed over 1 h continuous filtration in the recirculation mode. This indicates that Sb(III) oxidation is the main reaction process in the initial stage, then the Sb(V) can be adsorbed effectively by the loaded titanate nanowires. Furthermore, both Sb sorption kinetics and capacity increased with applied voltage due to enhanced electrostatic interactions and near surface transport by electromigration.

FIGURE LEGENDS:

Figure 1. Electroactive filtration apparatus. (1) is the anodic titanium ring connector to the anodic filter, (2) is the titanate-CNT anodic filter, (3) is the insulating seal, (4) is the cathodic CNT

filter, and (5) is the titanium ring connector to the cathodic filter.

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Figure 2. (A) FESEM and (B) TEM characterizations of the titanate-CNT filter. This figure has been modified from ref 19. Copyright 2019 Elsevier.

Figure 3. Changes of Sb species as a function of time. Experimental conditions: E = 2 V, $[Sb(III)]_0$ = 800 μg/L, flow rate =3 mL/min, pH_0 = 7 and 1 mM Na_2SO_4 electrolyte[19]. This figure has been modified from ref 19. Copyright 2019 Elsevier.

DISCUSSION:

The key to this technology is to fabricate an electroactive conductive and porous hybrid filter with high Sb-specificity. To do this, special care should be paid to the fabrication process. The amount of titanate nanowires need to be precisely controlled due to the "trade-off" effect between the filter's electrical conductivity and surface area.

In addition, it should be also noted that a proper applied voltage is necessary. Once the applied voltage is too high (e.g., >3 V), other competitive reactions, such as water splitting, may lead to the production of lot of bubbles (O_2 at the anode and H_2 at the cathode) at the electrode surface, which may block the active sites and, hence, contribute negatively to the Sb(III) removal performance.

The system stability in the long-term run is another issue of concern, since the accumulation of Sb-species on the filter's surface is inevitable. This requires periodically wash of the filter to regenerate the active surface sites (especially chemical wash).

Meanwhile, the cost of this electroactive titanite-CNT filter still needs to be considered. Although the price of CNTs has significantly decreased due to the progress of their production technology in the past decades, their prices are still far higher than that of activated carbon and other carbon materials that are widely used.

Furthermore, it is noteworthy that the current experimental results are mainly obtained from a laboratory-scale electrochemical filtration device. Further scaling up the device to enable practical large-scale environmental applications will be the focus of our subsequent study.

We have developed a continuous-flow filtration system for simultaneous Sb(III) adsorption and sequestration. The key to this technology is an electroactive titanite-CNT filter featured with electrochemical reactivity, small pore size, readily available active sites, and high Sb specificity. This study provides new insights for the rational design of flow-through systems towards the decontamination of Sb and other similar heavy metal ions.

ACKNOWLEDGMENTS:

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DISCLOSURE:

We have nothing to disclose.

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Figure 1

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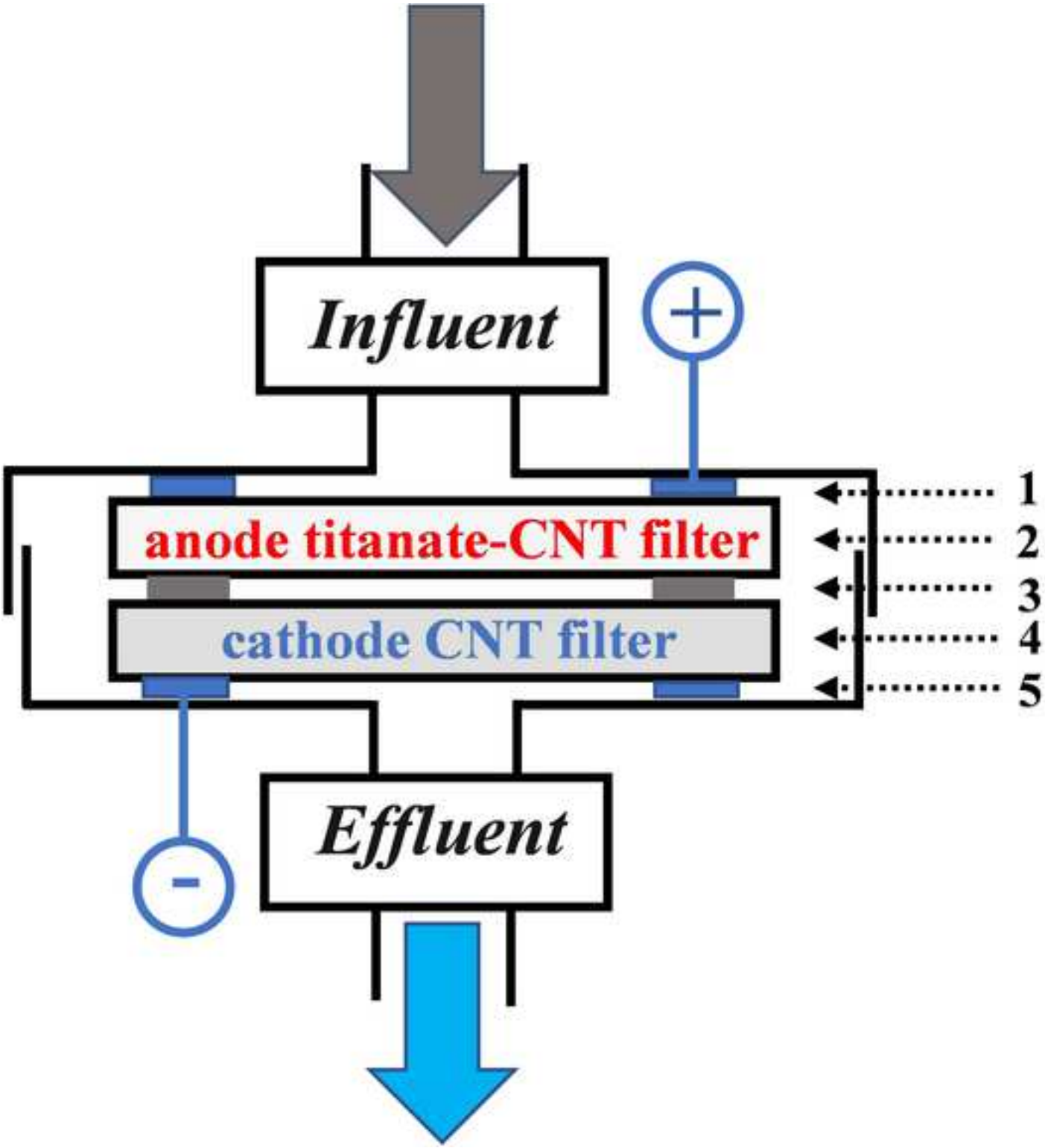

Figure 2

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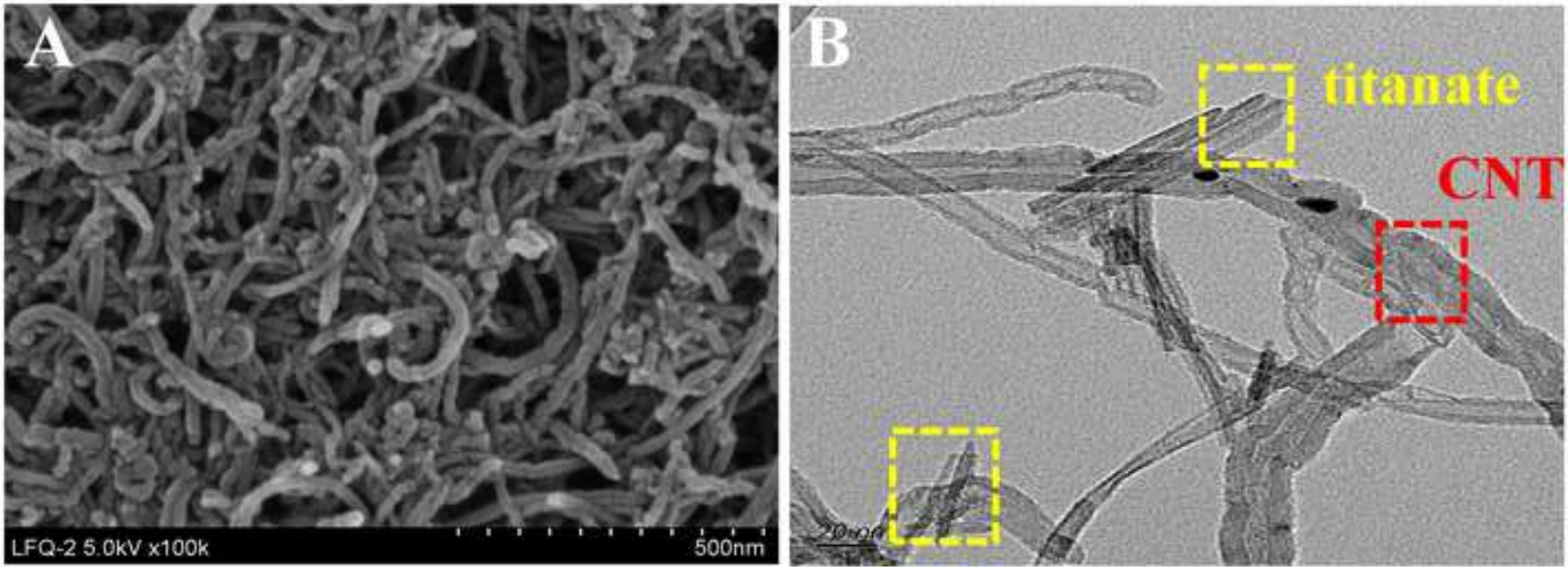
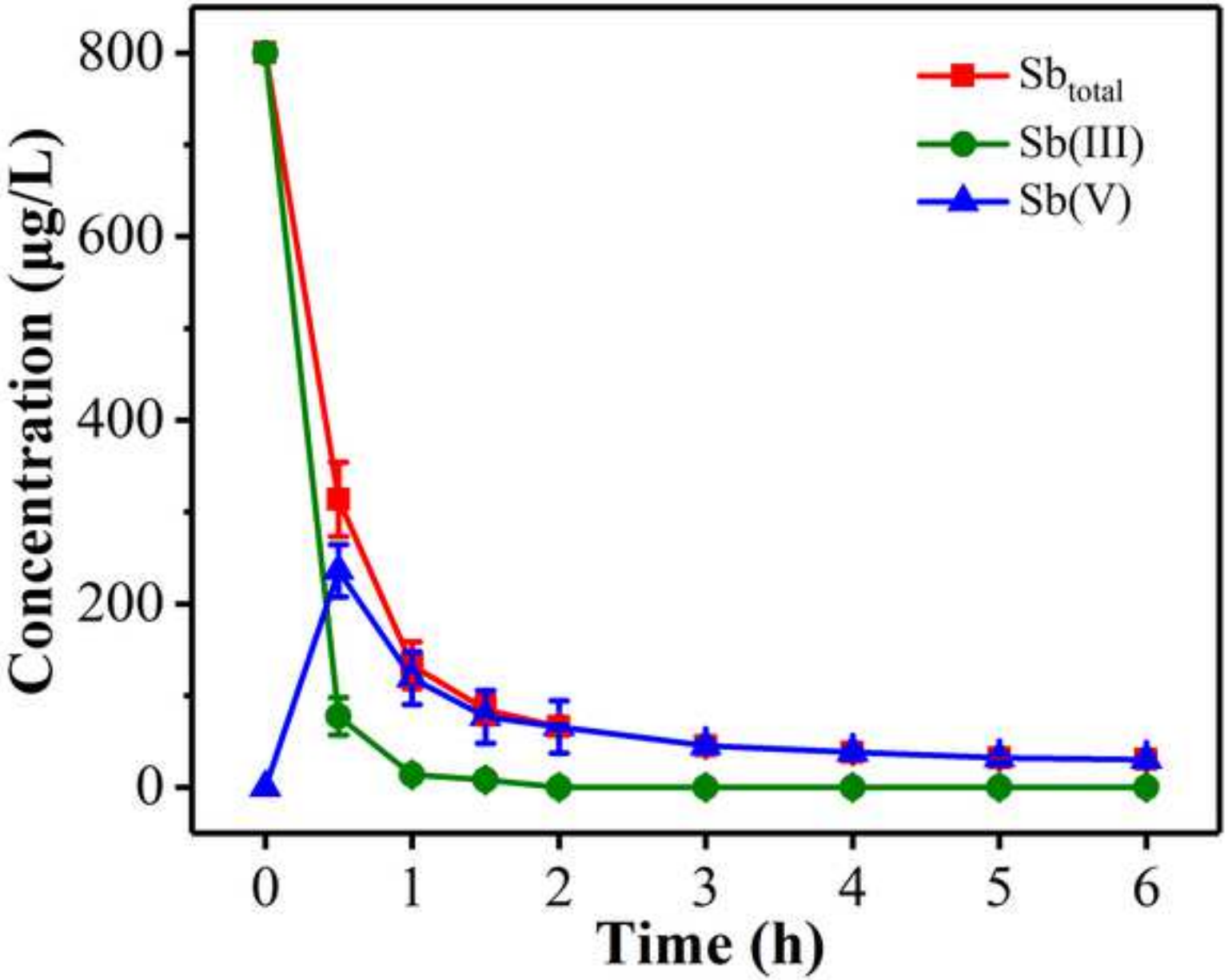


Figure 3



Name of Material/ Equipment	Company	Comments/Description
Atomic fluorescence spectrometer	Ruili Co., Ltd	
Carbon nanotubes (CNT)	TimesNano Co., Ltd	
DC power supply	Dahua Co., Ltd	
Ethanol, 96%	Sinopharm	
Hydrochloric acid, 36%	Sinopharm	Corrosive
L-antimony potassium tartrate	Sigma-Aldrich	Highly toxic
N-methyl-2-pyrrolidinone (NMP), 99.5%	Sinopharm	Highly toxic
Potassium hydroxide, 85%	Sinopharm	Corrosive
Peristaltic pump	Ismatec Co., Ltd	
Titanium dioxide powders	Sinopharm	

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Our Replies to the Editors' Comments

1. *Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.*

Reply: We have asked the professor Wolfgang Sand to double check the spelling and grammar issues.

2. *Please do not abbreviate journal titles for references.*

Reply: We have corrected this in the new version.

3. *Please remove the embedded Materials Table from the manuscript. All tables should be uploaded separately to your Editorial Manager account in the form of an .xls or .xlsx file.*

Reply: We have removed the embedded Materials Table.

4. *Step 2.2.5: Please add more details to your protocol steps. Please ensure you answer the "how" question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action.*

Reply: We have cited related references in the revised manuscript.

Revision:

Line 183,

"2.2.5) Determine the Sb_{total} and Sb(III) concentration with atomic fluorescence spectrometer technique[17]."

5. Please revise the Discussion to explicitly cover the following in detail in 3-6 paragraphs with citations:

a) Critical steps within the protocol

b) Any modifications and troubleshooting of the technique

c) Any limitations of the technique

d) The significance with respect to existing methods

e) Any future applications of the technique

Reply: We have supplemented the discussion section in the new manuscript.

Revision:

Line 226,

"The key to this technology is to fabricate an electroactive conductive and porous hybrid filter with high Sb-specificity. To do this, special care should be paid to the fabrication process. The amount of titanate nanowires need to be precisely controlled due to the "trade-off" effect between the filter's electrical conductivity and surface area. Furthermore, it is noteworthy that the current experimental results are mainly obtained from a laboratory-scale electrochemical filtration device. Further scaleup the device to enable practical large-scale environmental applications will be the focus of our subsequent study.

We have developed a continuous-flow filtration system for simultaneous Sb(III) adsorption and sequestration. The key to this technology is an electroactive titanite-CNT filter featured with electrochemical reactivity, small pore size, readily available active sites, and high Sb specificity. This study provides new insights for the rational design of flow-through systems towards the decontamination of Sb and other similar heavy metal ions."

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