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The study of lead removal from aqueous solution using an electrochemical method with a stainless steel net electrode coated with single wall carbon nanotubes

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HIGHLIGHTS

- A new electrode was used to remove lead form solution on this study.
- The mechanism of lead removal on electrode surface was studied.
- The effects of experimental parameters on lead removal were investigated.
- High lead removal was obtained in both synthetically and real wastewaters.
- The exhausted electrode can be easily regenerated using acid rinsed.

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ABSTRACT

The present work focused on the removal of lead from aqueous solution on a laboratory scale in a lab-made Plexiglas cell using an electrochemical technique. Electrodes of stainless steel net coated with single wall carbon nanotubes (SWCNTs@SSN) were used as both the anode and cathode. The mechanism of lead removal involved that lead ions were reduced and deposited on the surface of the cathode. The effects of various parameters on the percentage of lead removal were investigated, including electrochemical treatment time, solution pH, applied voltage, electrolyte concentration, and the initial lead concentration. Under optimal conditions with 90 min treatment, the lead removal efficiencies ranged from 97.2% to 99.6% when the initial lead concentrations varied from 20 mg dm^{-3} to 150 mg dm^{-3}. The SWCNTs@SSN electrodes could be easily regenerated. The removal results for lead, iron and aluminum in a factory wastewater using the proposed method showed that the method could be regarded as a potential technique for the treatment of industrial wastewater containing heavy metal ions.

1. Introduction

Pollution caused by heavy metal ions has attracted great concern because of their high toxicity and non-degradable characteristics. As one of the heavy metals, lead has special industrial significance, being employed in the battery, paint, pigments, ammunition, petrol, cable, alloy, steel, plastics and glass industries. Lead ranks first in the list of prioritized hazardous substances issued by the Agency for Toxic Substances and Disease Registry (ATSDR) of the United States [1]. Once lead is taken into the human body, it causes severe damage to the kidney, nervous system, reproductive system, liver and brain. Long time exposure to lead can induce sterility, abortion, stillbirths and neo-natal deaths [2]. In drinking water, a lead concentration lower than 50 μg dm^{-3} is considered safe by the World Health Organization (WHO) [3], whereas 15 μg dm^{-3} is the allowable limit regulated by the United States Environmental Protection Agency (USEPA) [4,5]. It is important to remove lead from wastewaters before they are discharged to the environment.

Numerous methods have been studied to remove lead from water and wastewaters, including chemical precipitation [6], adsorption [7], bioadsorption [8], ion exchange [9], membrane separations [10] and solvent extraction [11]. However, all these methods have certain disadvantages, among which are that chemical precipitation leads to the production of toxic sludge; solvent extraction is not suitable for effluents containing less than 1 g dm^{-3} of targeted heavy metals; and the ion exchange process is too expensive due to the high cost of synthetic resins and resin regeneration. To exploit new high efficient methods for lead removal is thus necessary.

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Electrochemical techniques can be used to treat wastewater and this particular application has been developed for several decades with some advantages, such as low cost and high convenience, and showing great potential in heavy metal wastewater treatment. Electrochemical reactions are induced by applying a current or potential onto a working electrode [12]. Electro-deionization uses electrically active media and applies an electrical potential to introduce ion transport, and can be operated in both batch and continuous modes. Conventional membrane electrodialysis involves alternating permselective cation and anion exchange membranes for separation of ions under an electric field. It allows only the cations or anions to permeate the corresponding membrane, so that ion diluting and ion concentrating compartments are separately created. The electrodialysis process has limited use in wastewater treatment, due to the problems associated with membranes, such as membrane fouling, scaling and concentration polarization [13,14]. The electrostatic shielding electrodialysis/electro-deionization process uses no ion exchange membranes but, instead, electrostatic shielding zones-ionic current sinks to create the alternating ion concentrating and ion diluting compartments [15,16]. Electrosorption and electrodeposition use electrodes to remove heavy metal ions from aqueous solutions, when an electrostatic field is applied on the surface of electrodes immersed in the aqueous electrolyte solution, and so the ions in the solution are forced to move to the oppositely charged electrodes [17]. On the surface of the electrodes, electrosorption and redox reactions may happen. As a result, the ions are removed from the solution.

In the electrochemical method, the electrode is one of the most important parts. During the past few years, conductive materials such as carbon aerogel [18], graphite [19], stainless steel [20], and vitreous carbon [21] have been applied as electrode materials in electrochemical reactors. On activated carbon and carbon aerogel electrodes some polymeric binders are often used to hold the carbon particles together. However, the use of these polymeric binders can induce occluding of the activated carbon surface and increase the electrical transfer resistance [22,23].

The electrochemical method has the unique feature of removing both inorganic and organic pollutants from aqueous solutions. Radionuclides, metal ions and anions are some typical target pollutants [24–26]. In our previous study, high percentage removals of Cr(VI) and total chromium were obtained with an electrochemical method [27]. The aim of the present work was to investigate the feasibility of using a stainless steel net (SSN) coated with single wall carbon nanotubes (SWCNTs), the (SWCNTs@SSN), as an electrode (SWCNTs@SSNE) for removal of the lead ions from an aqueous solution. An electrophoretic deposition method was adopted for the preparation of SWCNTs@SSNE. As described in our previous study [27], no polymeric binder was needed for preparing the SWCNTs@SSNE; therefore the problems existing in conventional activated carbon and carbon aerogel electrodes could be avoided.

2. Experimental

2.1. Materials and reagents

$NaNO_3$, HNO_3, NaOH, $Pb(NO_3)_2$ and N,N-dimethylformamide of analytical grade were supplied by the Sinopharm Chemical Reagent Co. (Shanghai, China). Ultra pure water (18.2 MΩ cm) was obtained from an ultra-pure water system (Unique-R30, Research Scientific Instruments Co., China). SWCNTs were purchased from the Institute of Solid State Physics of the Chinese Academy of Sciences (Hefei, China). The SSN (AISI304) of 40 meshes with wire diameter of 0.2 mm was purchased from a local store. Two types of water samples were used in the study. The stock lead solution ($1000\ mg\ dm^{-3}$) was prepared by dissolving $Pb(NO_3)_2$ in ultra pure water. The working lead solutions of desired concentration for the experiments were obtained by diluting the stock solution with ultra pure water. A real wastewater with pH 0.46 was collected from the effluent of a local wiring factory located at Quanzhou, Fujian. The concentrations of heavy metals in the wastewater are listed in Table 1.

Table 1
Characteristics of a factory effluent (mg/L).

Metal	Pb	Na	Mg	Al	Fe	Ca
Concentration	12.76	3.46×10^4	45.10	13.92	21.75	46.50

2.2. The electrolysis cell and preparation of SWCNTs@SSNE

The experiment for parameter optimization was carried out at a laboratory scale in a home-made Plexiglas cell. The electrolysis cell and the preparation of SWCNTs@SSNE are described in our previous work [27]. Briefly, the dimensions of the electrolysis cell were 8.0 cm in length, 6.5 cm in height, and 4.0 cm in width. The applied voltage was supplied by an electrochemical analytical instrument (CHI630B, Shanghai Chenhua Instrument Co., China). A saturated calomel electrode (SCE) was selected as the reference electrode, and two SWCNTs@SSNEs were used as cathode and anode. A magnetic stirrer (JBZ-14, Shanghai Kang-Yi Instruments Co., China) was used to maintain the electrolyte homogenously with the rotation speed of 240 rpm.

In the process of preparing the SWCNTs@SSNE, the SSN was cut into 6.5 cm × 4.5 cm pieces and rinsed with sulfuric acid solution (1:10, v/v). One gram of SWCNTs with pore size 1–2 nm, length 5–30 μm, and specific surface area more than $380\ m^2\ g^{-1}$, was refluxed in a mixed solution of nitric and sulfuric acid (1:3, v/v) at 130 °C for 45 min. After the mixture was brought to pH 7.0 with pure water and centrifuged with 5000 rpm, the SWCNTs were dried in a constant temperature blast dryer (DHG-9146A, Shanghai Jing-hong Instrument Co., China). Then the treated SWCNTs were ultrasonically dispersed in dimethylformamide and a suspended solution containing SWCNTs was obtained. Two pieces of clean SSN with dimension 6.5 cm × 4.5 cm were immersed in parallel into the suspension of SWCNTs as electrodes. A constant voltage of 40 V was applied to deposit SWCNTs on the SSN anode. The SSN anode was dried at 120 °C to remove the solvent, and it was then ready for use as the SWCNTs@SSNE. The SWCNTs remained tightly attached on the surface of the stainless steel, and no break or crack was observed even after being immersed in water, acid and alkaline solutions, respectively.

The reason why SWCNTs were deposited on the SSN anode was because some oxygenated groups, such as carboxyl and hydroxyl, were introduced onto the surface of the SWCNTs during the acid treatment processing of the SWCNTs [28]. The carboxyl and hydroxyl groups were negatively charged, and thus the acid treated SWCNTs were also negatively charged due to the occurrence of carboxyl and hydroxyl groups. When a constant voltage of 40 V was applied on the two pieces of SSN immersed in the suspension of SWCNTs, the SWCNTs would move towards the SSN as the anode under electrical attraction.

2.3. Electrochemical treatment process

The pH of the solution was adjusted, using a pH meter (UB-7, Denver Instrument, USA), with NaOH or HNO_3 solution. The experiments were carried out at pH 6.50, voltage −2.0 V (vs. SCE), current about 0.01 A, solution volume $180\ cm^3$, electrode dimension 6.5 cm × 4.5 cm and distance 3 cm, electrolyte $NaNO_3$