



Efficient selenium(IV) detection and removal from water by tailor-made novel conjugate adsorbent



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ABSTRACT

Selenium occurs naturally in the environment and is toxic at elevated concentrations, which has been a challenging issue for environmental scientists. This work was devoted to the detection and removal the selenium (Se(IV)) from aqueous solutions using organic ligand an immobilized conjugate adsorbent. The organic ligand of (3-(3-(methoxycarbonyl) benzylidene)hydrazinyl) benzoic acid was synthesized and indirectly immobilized onto the mesoporous silica. The adsorbent was applied to detect and remove Se(IV) in aqueous solutions at optimum conditions. This adsorbent exhibited high surface area-to-volume ratios and pores were uniform nanostructures. The adsorbent permitted rapid Se(IV) capturing detectable by naked eye observation. The limit of detection was 2.02 $\mu\text{g/L}$ by the proposed method of the conjugate adsorbent. The Se(IV) capture system was optimized by varying the parameters of contact time, pH, initial concentration, competing ions and elution operations. The pH effect played an important role and the solution pH ranging from 0.5 to 7.0 was investigated. The data confirmed that acidic pH enhanced the Se(IV) sorption on the conjugate adsorbent resulting in an increased sorption efficiency percentage of Se(IV) to >98% at pH 1.50. The data were also fitted to the Langmuir isotherm, and the maximum sorption capacity was 93.56 mg/g. The introduction of diverse ions was individually added to solution, and these had no significant effect on the Se(IV) sorption by the adsorbent. The 0.20 M NaOH was used an eluent in the elution operation, and the adsorbent was used in many cycles without loss of cage cavities. Therefore, the conjugate adsorbent has shown favorable capturing ability in water samples, and this could be used as efficient adsorbents in potential practical applications for Se(IV) detection and removal from wastewater.

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1. Introduction

Selenium is an essential micronutrient for humans and animals at low concentrations, but highly toxic at elevated concentrations. Selenium is also an important material used in many devices such as photoelectric devices, pigments, solar batteries, semi-conductors, colorization and decolorization of glasses, photosensitive drums and as a catalyst in the synthesis of urea and urethane [1,2]. Thereby, a considerable amount of selenium-contaminated wastewater is generated. Selenium also exists in radioactive wastes as radionuclides (⁷⁵Se, ⁷⁹Se) in spent nuclear fuels. The ⁷⁹Se (half-life, 6.5 × 10⁴ years) has significant effects on the cumulative dose of radioactivity because it is one of the most contributing isotopes to global radioactivity that could be

potentially released to the biosphere term [3,4]. Also, an elevated amount of selenium is found in the effluents from oil refineries, thermal power stations, industrial wastewater, and combustion from fossil fuels and agricultural drainage water from irrigation [5,6]. Therefore, selenium pollution arises from both natural and anthropogenic sources. Selenium is highly toxic and causes hair and fingernail loss, damage to the peripheral nervous system, fatigue and irritability and damage to kidney and liver tissue as well as the nervous and circulatory systems [7]. Selenium exists in four oxidation states such as elemental (Se⁰), selenide (Se²⁻), (SeO₃²⁻, Se(IV)) and selenate (SeO₄²⁻, Se(VI)) [8]. However, Se(IV) and Se(VI) are dominant in aqueous systems. Moreover, Se(IV) is more toxic than the Se(VI) [9]. In aqueous effluents, selenium is usually present in the range from 0.1 to 20 ppm. These levels are too high, and it is important to treat such effluents before discharge. The World Health Organization (WHO) currently sets the maximum permissible limit in drinking water is 10 $\mu\text{g/L}$ [10], and USEPA allow a maximum contaminant level of 50 $\mu\text{g/L}$ [11].

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Several methods have been developed to determine the presence of selenium, such as inductively coupled plasma atomic emission spectroscopy (ICP-AES), graphite furnace atomic absorption spectroscopy (GF-AAS), differential pulse cathodic stripping voltammetry (DPCSV) differential pulse polarography (DPP) combined with cathodic stripping voltammetry (CSV), ion chromatography and neutron activation analysis [12–16]. Many of these are time consuming, costly, cumbersome, unsuitable in developing or less-developed countries and have high training requirements [17]. Therefore, there is increasing demand to develop cost-effective, easy to use, reliable and robust methods to detect the selenium in water samples. Recent analytical interest has focused on developing optical adsorbents that can be reused and do not involve sample destruction. Colorimetric adsorbents based on decorated mesoporous particles have attracted much attention for their conveniences of visual observation and simple operations. In this connection, the solid based sensors have great advantages in terms of selectivity, sensitivity and ease of use for scientists [17,18]. Therefore, designing effective methods that allow direct analysis of the samples by the naked eye without using highly sophisticated instruments is still required. So, it is a big challenge to develop efficient solid based optical materials to perform the sequential determination and removal of selenium from water samples.

There are various treatment methods reported for selenium removal from contaminated water such as chemical precipitation, evaporation, solvent extraction, coagulation/flocculation, solidification/stabilization, extraction with chelating agents, reverse osmosis, nanofiltration, ion exchange, electrochemical operation, cementation, lime softening, granular activated carbon and powdered activated carbon and adsorption [19–24]. However, most of the separation methods still suffer from low selectivity and sensitivity that limit their practical application. The adsorption process is one of the most promising techniques for selenium removal from water. Considerable work has been reported for various adsorbents such as metal oxide adsorbents and biomass such as green algae, modified rice husk and peanut shell [25–28]. However, many of these adsorbents are unable to uptake the selenium in the presence of competing anions such as Cl^- , NO_3^- , CO_3^{2-} , SO_4^{2-} and phosphate. The organic–inorganic based conjugate materials are promising candidates to take up the specific ion at optimum conditions in the second generation of a solid–liquid phase extraction system. The conjugate materials are superiors because of their high surface area, large pore volumes, and lack of internal diffusion resistance. Therefore, the specific functional group containing ligand immobilized materials are attractive due to their complex formation ability, long mechanical stability, high sorption efficiency and extreme selectivity. Here, we prepared functional group ligand immobilized new class conjugate adsorbent for efficient detection and removal of Se(IV) from wastewater.

In this work, we have prepared organic–inorganic materials based conjugate adsorbent for efficient Se(IV) detection and removal from water solutions. For preparation of the new class conjugate adsorbent, a specific functional group of organic ligand ((3-(3-(methoxycarbonyl)benzylidene)hydrazinyl) benzoic acid (MBHB)) was prepared and successfully incorporated into mesoporous silica by non-specific interaction according to the indirect immobilization approach. The mesoporous silica exhibited highly ordered structure, and the pores were well arranged to use as an excellent substrate for combination with an organic ligand such as MBHB. Therefore, the MBHB was embedded onto mesoporous silica, namely non-specific interaction via hydrogen bonding, Van der Waals forces and reversible covalent bonds. The conjugate adsorbent was triggered by Se(IV) ions at optimum pH conditions, and the signal intensity was transduced upon color optimization. The conjugate adsorbent has high selectivity toward Se(IV) ions over competitive anions such as Cl^- , NO_3^- , CO_3^{2-} ,

SO_4^{2-} and phosphate and is also able to detect the Se(IV) directly based on the colorimetric change by optical visualization. Several experimental parameters such as solution pH, color optimization, limit of detection and quantification, adsorbent dosage, contact time, Se(IV) sorption capacity, the presence of competing ions and elution/regeneration behavior were evaluated and optimized systematically. Moreover, the developed adsorbent is cost-effective and suitable to the large-scale treatment for Se(IV) capturing in contaminated water samples.

2. Materials and methods

2.1. Materials

All materials and chemicals were of analytical grade and used as purchased without further purification. Tetramethylorthosilicate (TMOS), F108 ($\text{EO}_{141}\text{PO}_{44}\text{EO}_{141}$) and methyl 3-formylbenzoate were obtained from Sigma–Aldrich Company Ltd. USA. Selenium dioxide (SeO_2) was purchased from Sigma–Aldrich. The Se(IV) stock solutions (200 mg/L) were prepared by dissolving SeO_2 in Millipore water. For pH adjustments in detection systems, buffer solutions of 3-morpholinopropane sulfonic acid (MOPS) and sodium acetate were procured from Dojindo Chemicals, Japan, and KCl, HCl and NaOH from Wako Pure Chemicals, Osaka, Japan. The other cations and anions salts were also purchased from Wako Pure Chemicals, Osaka, Japan. Ultra-pure water prepared with a Millipore Elix Advant 3 was used throughout in this work.

2.2. Synthesis and characterization of MBHB ligand

The structure and major steps for the preparation of the (3-(3-(methoxycarbonyl) benzylidene)hydrazinyl)benzoic acid (MBHB) ligand is shown in Scheme 1. The preparation methods are reported elsewhere [29a]. However, the preparation is described briefly for the reader's understanding. Here, the MBHB was prepared by the reaction of methyl 3-formylbenzoate (1 mol) and 3-hydrazinylbenzoic acid (1 mol) in ethanol and a small amount of acetic acid. The resultant mixture was then refluxed for 4 h and left to cool at room temperature. The solid material formed upon cooling was collected by suction filtration. The separated product was recrystallized with a mixed solvent of dichloromethane/methanol 1/1.

2.3. Instruments and analyses

The NMR spectra was obtained on a Varian NMR System 400 MHz Spectrometer. The N_2 adsorption–desorption isotherms were measured using the 3Flex analyzer (Micromeritics, USA) at 77 K. The pore size distribution was measured from the BJH adsorption. Mesoporous inorganic silica was pre-treated at 100 °C for 3 h under vacuum until the pressure was equilibrated to 10^{-5} Torr before the N_2 isothermal analysis. The specific surface area (S_{BET}) was measured by using multi-point adsorption data from the linear segment of the N_2 adsorption isotherms using Brunauer–Emmett–Teller (BET) theory. Transmission electron microscopy (TEM) was obtained by using a JEOL (JEM-2010) and operated at the accelerating voltage of the electron beam at 200 kV. The TEM samples were prepared by dispersing the powder particles in ethanol solution using an ultrasonic bath and then dropped on copper grids. The absorbance spectrum was measured by UV–Vis–NIR spectrophotometer (Shimadzu-3700). The metal concentrations were measured by ICP-AES (SII NanoTechnology Inc.). The ICP-AES instrument was calibrated using five standard solutions containing 0, 0.5, 1.0, 1.5 and 2.0 mg/L (for each element), and the correlation coefficient of the calibration curve was

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