



## Research article

## Removal of uranium(VI) ions from aqueous solutions using Schiff base functionalized SBA-15 mesoporous silica materials

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## ARTICLE INFO

## Article history:

Received 19 June 2015

Received in revised form

5 December 2015

Accepted 7 December 2015

Available online 22 December 2015

## Keywords:

Uranium(VI)

Adsorption

Solid phase extraction

Functionalized SBA-15

Decontamination

Water samples

## ABSTRACT

Functionalized SBA-15 mesoporous silica particles, bearing *N*-propylsalicylaldehyde and ethylenediaminepropylsalicylaldehyde Schiff base ligands, abbreviated as SBA/SA and SBA/EnSA respectively, were prepared and characterized by FT-IR, elemental analysis, TGA, XRD, TEM and SEM techniques. The potentials of these adsorbents were examined by using them in solid phase extraction of U(VI) ions from water samples. It is shown that 20 mg of SBA/SA or SBA/EnSA can remove rapidly (~15 min) and quantitatively uranium(VI) ions from 10 to 200 mL of water solutions (pH 4) containing 0.2 mg of the ions, at 25 °C. The adsorbed ions were stripped by 1 mL of dilute nitric acid solution (0.1 mol L<sup>-1</sup>). It means that the studied adsorbents are able to be used for removal and concentration of uranyl ions. This allowed achieving to a concentration factor of 200 for uranyl ions. The variation in the ionic strength in the range 0–1 mol L<sup>-1</sup> did not affect the extraction efficiencies of the adsorbents. The adsorbents showed selective separation of uranyl ions from Cd<sup>2+</sup>, Co<sup>2+</sup>, Ni<sup>2+</sup>, Mn<sup>2+</sup>, Cr<sup>3+</sup>, Ba<sup>2+</sup>, Fe<sup>3+</sup> and Eu<sup>3+</sup> ions. Thermodynamic investigations revealed that the adsorption of uranyl ions by the adsorbents was spontaneous and endothermic. The Langmuir model described suitably the adsorption isotherms. This model determined the maximum adsorption capacity of the adsorbents SBA/SA and SBA/EnSA as 54 and 105.3 mg uranyl/g adsorbent, respectively. The kinetics of the processes was interpreted by using Pseudo-second-order model.

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## 1. Introduction

Hexavalent uranium plays a crucial role in nuclear energy production industries. Besides, it is one of the most environmentally important heavy metals due to its toxicity, mobility and radioactivity (Sprynskyy et al., 2010). Uranium can be a menace for human life and it needs many years for reducing its radioactivity to the safe level. It results in progressive renal damage and causes to kidney failure and death (Domingo, 2001; Thiebault et al., 2007; Majdan et al., 2010a). Thus, the removal of uranium from mine tailings effluent, waste waters and environmental waters is of great importance from both keeping human health and environmental protection points of view (Han et al., 2007).

A variety of techniques have been applied for the removal of U(VI) from water solutions. Among these techniques are evaporation, ion exchange, solvent extraction, transport through

membranes and precipitation (Deliyanni et al., 2003; Mellah et al., 2006). These techniques are generally ineffective for removal of trace amount of pollutants and require high capital investment (Ghasemi et al., 2011). Solid-phase extraction methods are also among these techniques bearing some advantages such as simplicity, reliability, ability to get high enrichment factor and low consumption of organic solvents.

Various natural, organic and inorganic materials have been examined as adsorbent for uranium(VI) ions removal from aqueous solutions. As instance, Chitosan (Humelnicu et al., 2011), marine algal biomass (*Padina* sp.) (Khani, 2011), silica materials (Venkatesan et al., 2004; Cyriac and Balaji, 2010), P(4-vinyl pyridine) hydrogel (Ozay et al., 2011), humic acid (Khalili and Al-Banna, 2015), ion imprinted polymer (Gladis and Rao, 2004; Preetha et al., 2006), anion exchange resins (Semnani et al., 2012), magnetic composite (Fan et al., 2012), carboxylate-functionalized poly(hydroxyethylmethacrylate)-grafted lignocellulosics (Anirudhan et al., 2009), nanospheres (Milja et al., 2011; Pandey et al., 2011), clay (Majdan et al., 2010b), coir pith (Parab et al., 2005), zeolites (Aytas et al., 2004; Kilincarslan and Akyil, 2005; Han et al., 2007),

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cork biomass (Psareva et al., 2005), goethite (Missana et al., 2003), activated carbon (Mellah et al., 2006), graphene oxide-activated carbon (Chen et al., 2013), cement (Awwad and Daifullah, 2005), akaganeite (Yusan and Akyil, 2008), and pyrolyzed tea and coffee wastes (Aly and Luca, 2013) are among the adsorbents used for this aim.

Recent developments in preparation of effective adsorbents result in preparation of mesoporous materials. These materials possess high surface areas, uniform pore structures, excellent mechanical stabilities and high adsorption capacities. SBA-15 based adsorbents, as common mesoporous molecular sieves, are an important group of materials used in solid phase extraction processes (Bruzzoniti et al., 2007; Hulea et al., 2010; Liu and Du, 2011; Degirmenci et al., 2011; Sayari et al., 2011; Park et al., 2011; Hami Dindar et al., 2015a; Hami Dindar et al., 2015b). Liu et al. reported application of SBA-15 supported Pb(II)-imprinted polymer as adsorbent for selective removal of lead ions from aqueous solutions (2011). The elimination of copper ions from water samples by tetrakis(4-carboxyphenyl)porphyrin functionalized SBA-15 has been studied by Park et al. (2011). Hulea et al. found that the 1-furoyl thiourea functionalized SBA-15 can effectively remove Hg(II) from aqueous solutions (2010).

Functionalized mesoporous materials have gained also great attention for removal of uranium ions (Vidya et al., 2002, 2004; Benhamou et al., 2009; Yousefi et al., 2009; Lee et al., 2011; Liu and Du, 2011). Yousefi et al. (2009) studied the solid phase extraction of U(VI) using 5-nitro-2-furaldehyde modified mesoporous silica. Application of monoamide resins with porous silica support for selective recovery of U(VI) from nitric acid media has been investigated by Nogami et al. (2007; 2010a; 2010b).

The present paper concerns on the investigation of ability and applicability of two SBA-15 mesoporous silica materials functionalized by Schiff base ligands *N*-propylsalicylaldehyde and ethylenediaminepropylsalicylaldehyde (abbreviated as SBA/SA and SBA/EnSA, respectively) for solid phase extraction of U(VI) ions from water solutions. To the best of our knowledge, there is only one report on the application of amino-functionalized SBA-15 for removal of uranium(VI) ions from aqueous solutions (Liu et al., 2012). The effect of aqueous solution pH, adsorbent dose, shaking time, initial concentration of uranyl, aqueous solution volume, ionic strength, temperature and the presence of other ions, on the extraction has been verified and discussed. The thermodynamic parameters of the processes were evaluated. The Langmuir, Freundlich and Dubinin–Radushkevich isotherm models have been examined for describing the equilibrium data. In order to study the kinetics of the processes, pseudo first-order and pseudo second-order equations were applied. The presented procedures were examined for the removal of U(VI) ions from water samples.

## 2. Experimental

### 2.1. Reagents and materials

All the chemicals used were analytical grade and were furnished by Merck, Fluka or Acros chemical companies. Poly(ethyleneglycol)-block-poly(propyleneglycol)-block-poly(ethyleneglycol) called surfactant P123, tetraethoxysilane (TEOS), aminopropyltriethoxysilane (APTS), (2-aminoethyl-aminopropyl)trimethoxysilane (TPED), salicylaldehyde, toluene and ethanol were used for preparation of the adsorbents. A stock solution of 1000 mg L<sup>-1</sup> uranium(VI) was prepared by dissolving appropriate amount of UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O in deionized water containing 0.01 mol L<sup>-1</sup> nitric acid. The working solutions were prepared by diluting the stock solution with

deionized water. The pH adjustments were performed by addition of diluted nitric acid or sodium hydroxide solutions.

### 2.2. Instrumentation

An inductively coupled plasma optical emission spectrometry (ICP OES, Spectro Genesis, Germany), equipped with a peristaltic pump and a cyclonic spray chamber with a concentric nebulizer, was used for determination of uranium ions. The purity of argon gas used in the analysis was 99.999%. FT-IR spectra were recorded on a Thermo-Nicolet-is10 FT-IR spectrometer. The X-ray powder diffraction pattern was recorded on a Bruker XRD diffractometer (B8-Advance) using Cu K $\alpha$  radiation in the range 0.5–10 $^{\circ}$  2 $\theta$ . The morphology of the mesoporous materials was obtained by scanning electron microscopy (SEM). The shape and size of the pores of the mesoporous materials were investigated by transmission electron microscope (TEM). Thermogravimetric analyses were done by using a NETZSCH STA 409 PC/PG instrument. Solid-liquid phase separation was assisted by a centrifuge Heraeus Labofuge 300. A Metrohm pH meter (model 780) with a combined pH electrode was used for pH measurements. Deionized water (18.2 M $\Omega$ ) was used throughout performing the experiments and was provided by a water purification system (Zolalan, M-UV-3+, Iran). An Elmasonic ultrasonic bath which worked at 37 kHz and 138 W power, was used for ultrasonic irradiations.

### 2.3. Preparation of the adsorbents

A schematic representation of the synthesis of the adsorbents is given in Fig. 1.

#### 2.3.1. Preparation of SBA-15 skeleton

SBA-15, as skeleton of the studied adsorbents, was prepared based on a previously reported procedure (Zhao et al., 1998; Hongping et al., 2002). In a typical synthesis batch, 4 g of triblock copolymer surfactant (P123) as template was dissolved in 30 mL deionized water and 120 mL hydrochloric acid (2 mol L<sup>-1</sup>). TEOS (9 g) was added to the reaction mixture and it was stirred for 8 h at 40  $^{\circ}$ C. The resulting mixture was transferred into a Teflon-lined stainless steel autoclave and kept at 100  $^{\circ}$ C for 20 h. After cooling down, the product was filtered, washed with water and dried at 70  $^{\circ}$ C. The template was removed by calcinating the prepared material at 550–600  $^{\circ}$ C for 6 h. The XRD of the mesoporous silica SBA-15 showed three diffraction peaks that can be indexed as hexagonal symmetry. The  $d_{100}$  value corresponds to the distance between two successive walls (that is, one pore diameter plus a wall thickness). Diffraction peaks at the below  $2^{\circ}$  corresponding to the (1 0 0), (1 1 0) and (2 0 0) are recognized from the XRD pattern of SBA-15. The observed diffraction peaks agree with the 2 D-hexagonal SBA-15. In the FT-IR spectrum of SBA-15, the bands at  $\sim$ 800 and  $\sim$ 1100 cm<sup>-1</sup> is attributed to Si–O–Si bond vibrations. The small band at about 960 cm<sup>-1</sup> is related to Si–OH bond. The Si–OH groups are appeared by a broad absorption band in the region 3000–3700 cm<sup>-1</sup>.

#### 2.3.2. Preparation of SBA-15/PrNH<sub>2</sub>

The calcinated SBA-15 particles (1 g) was added to 100 mL of (3-aminopropyl)triethoxysilane solution (0.01 mol L<sup>-1</sup>) in dry toluene. This mixture was refluxed for 10 h. The product was filtered, and then dried under vacuum. CHN analysis showed the prepared mesoporous loaded with 0.62% of nitrogen (Hami Dindar et al., 2015a).

#### 2.3.3. Preparation of SBA-15/PrEn

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