Separation and Purification Technology 210 (2019) 491–496

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Tailored synthesis of SBA-15 rods using different types of acids and its application in adsorption of uranium

Hui Dan, Luyu Chen, Qiang Xian, Facheng Yi*, Yi Ding*

Key Subject Laboratory of National Defense for Radioactive Waste and Environmental Security, Southwest University of Science and Technology, Mianyang 621010, PR China

ARTICLE INFO

Keywords:
SBA-15 rods
Acid type
Powder quartz
Uranium
Adsorption

ABSTRACT

Monodisperse SBA-15 rods with tunable size, synthesized by a facile and low-cost route using different types of acids (HCl (monobasic acid), H_2SO_4 (binary acid) and H_3PO_4 (ternary acid)) as media and inexpensive powder quartz as silica source without any additives, was developed as an absorbent to extract uranium from radioactive waste water. The effect of the type of acid on the structure and morphology of the obtained mesoporous SBA-15 was investigated. SBA-15 rods with varying lengths (1082–1354 nm) and diameters (279–361 nm) were tailored by simply using different types of acids. The number of H in acid molecular was identified to be the key factor affecting the structure and morphology of the SBA-15 rods. It was found that the length and diameter of these SBA-15 rods increased with increase of the number of H in acid molecular. Furthermore, the adsorption behavior of uranium on these SBA-15 rods was investigated and the results showed that all these SBA-15 rods demonstrated rapid (30 min to reach equilibrium) and high capacity (235 mg g^{-1}) for adsorption of uranium. This work demonstrated that these SBA-15 rods prepared with different types of acids from powder quartz exhibited improved performance for the extraction of uranium.

1. Introduction

Uranium, a chemical toxic and radioactive nuclide that enters the environment through activities associated with the nuclear industry, can cause surface and ground water contamination, which results in serious problems for human health and damage of bio-organisms [1]. Uranium in radioactive waste water is not only the main radioactive pollution nuclide, but also the raw materials of the nuclear industry. Hence, effective extraction of uranium not only reduces radioactive hazards, but also alleviated uranium resource shortage. Several methods have been employed to remove uranium from aqueous such as chemical precipitation [2–4], ion exchange [5,6], membrane processes [7,8], solvent extraction [9] and adsorption [10–15].

Among those methods, adsorption has attracted much attention due to its high efficiency, high concentration factor and easy operation. Subsequently, a number of absorbents including carbon nanotubes [16], mental oxides [17], clay [18], chitosan [19] and mesoporous silica [20–24] have been employed for the removal of uranium from aqueous solutions. Recently, several new types of adsorbents were developed to remove uranium such as metal-organic framework-based materials [25], polymer [26], layered double hydroxide-based nanomaterials [27] and boron nitride-based materials [28]. Among these absorbents, SBA-15, a mesoporous silica with high surface area, large and uniform pore size, thick pore walls and excellent hydrothermal stability, has great potential applications in the extraction of uranium from radioactive waste water.

Since the discovery of silica based mesoporous materials [29], considerable efforts have been made to understand their synthesis process in order to control their macro morphologies and micro structures. Mesoporous silica with desired morphology and structure usually exhibits excellent application performance. For instance, SBA-15 rods with short and straight pore channels possess much improved immobilization abilities for enzyme over that of conventional morphology (fiber like) SBA-15 [30,31]. Up to now, mesoporous silica with various morphologies, such as fiber, rod, platelet, sphere and film have been prepared through using additives or varying the synthetic parameters [32–37]. Generally, in order to achieve the controllable synthesis of mesoporous materials, employing additives is a necessary strategy. However, the introduction of additives will make self-assembly system interactions more complex between species and species. Therefore, mesoporous materials with diverse morphologies or disordered microstructure resulted usually when additives are employed [38].

Tetraethyl orthosilicate (TEOS), an organic compound, is usually used as the silica source to synthesize mesoporous silica materials

* Corresponding authors.
E-mail addresses: yfc66@163.com (F. Yi), dingyi2279@126.com (Y. Ding).

https://doi.org/10.1016/j.seppur.2018.08.039
Received 3 July 2018; Received in revised form 7 August 2018; Accepted 21 August 2018
Available online 23 August 2018

[39,40]. Thus, it is harmful to environment. Furthermore, a high cost is resulted, which may limit its application severely. It is a good inspiration for us that there have been several reports on the preparation of silica materials from cheap rice husk [41,42]. An alternative silica source, powder quartz is cheap mineral and has high silica content. It may be a good candidate for synthesizing mesoporous silica materials.

As known, the acid synthesis route is well developed because a rich morphology is achievable in acidic conditions [43]. It was found that mesoporous silica materials with various morphologies can be obtained by adjusting acidity [44,45]. In this route, the acid plays a key role as a catalyst for hydrolysis and condensation of silica source, and has been known to have strong effects on the morphology and structure of mesoporous silica materials [46,47]. Among the various acids, HCl (monobasic acid) is often used as a catalyst for hydrolysis and condensation of silica source when the acid synthesis route is employed. In particular, as reported by Ding et al. [48] that SBA-15 rods with various lengths (0.6–3.0 μm) and widths (0.26–0.40 μm) can be obtained by varying the HCl concentration in the range of 0.5–2.5 M. A possible mechanism for the effect of the HCl concentration on the growing manner of the rod-like particles was proposed. However, up to now, the other types of acids (eg. H_2SO_4 (binary acid), H_3PO_4 (ternary acid)) are seldom used as media in the synthesis of mesoporous silica materials. Therefore, the effects of the type of acid on the morphology and structure of mesoporous silica are not well known.

From the above options, in this work, we present a novel and simple approach for synthesizing ordered mesoporous silica SBA-15 rods using different types of acids (HCl, H_2SO_4 and H_3PO_4) as media and inexpensive powder quartz as silica source without additives. And the obtained SBA-15 materials were developed as an absorbent to extract uranium from radioactive waste water. The effects of the type of acid on the morphology and structure of the obtained mesoporous silica were investigated. Moreover, the adsorption performance of uranium on these mesoporous silica materials was evaluated. The results demonstrated that these obtained SBA-15 rods exhibited improved performance for the extraction of uranium and that it has the potential for practical applications. The schematic diagram of research idea in this work was shown in Fig. 1.

2. Materials and methods

2.1. Materials

Triblock copolymer Pluronic P123 ($EO_{20}PO_{70}EO_{20}$, M_{av} = 5800) was purchased from Sigma-Aldrich. Hydrochloric acid (HCl), sulphuric acid (H_2SO_4), phosphoric acid (H_3PO_4) and sodium carbonate (Na_2CO_3) were purchased from Kelong Chemical Reagent Factory (Chengdu, China). Uranyl nitrate hexahydrate $UO_2(NO_3)_2{\cdot}6H_2O$ was obtained from the China Academy of Engineering Physics. Powder quartz (350 mesh, 99.5 wt% SiO_2) was obtained from Sichuan Xinju Mineral Resource Development Co., Ltd (Chengdu, China). All of the reagents were of AR grade and used without further purification. A standard stock solution (500 mg/L) of U(VI) was prepared by dissolving the appropriate amounts of $UO_2(NO_3)_2{\cdot}6H_2O$ in deionized water. The various concentrations of U(VI) solutions were prepared by diluting the stock solution.

2.2. Preparation of mesoporous SBA-15

The powder quartz and sodium carbonate were mixed with the molar ratio of 1:1, and then heated at 1450 °C for 2 h to obtain solid sodium silicate, followed by dissolving it using deionized water in high-pressure reactor to obtain sodium silicate solution. Mesoporous SBA-15 silica was synthesized using the obtained sodium silicate solution as silica source. In a typical synthesis, 2.4 g P123 was dissolved in 120 mL of 2 M different types of acids (HCl, H_2SO_4 and H_3PO_4) aqueous solution, which was stirred at 40 °C to get a transparent solution. Subsequently, calculated sodium silicate solution was slowly added to the solution. The mixture was stirred for 1 min. Consequently, the mixture was kept under static conditions at the 40 °C for 20 h. After that the mixture was transferred to an autoclave for hydrothermal treatment at 100 °C for another 24 h. Finally the resulting precipitate was filtered, washed carefully with distilled water, air dried. P123 was removed by calcination at 550 °C for 6 h in air. The samples obtained from HCl, H_2SO_4 and H_3PO_4 were designated as H1, H2 and H3, respectively, while keeping other synthesis parameters constant.

2.3. Adsorption of uranium

The adsorption experiments were carried out using the batch method with initial concentrations of U(VI) ranging from 20 to 200 mg L^{-1}. The pH was adjusted using diluted nitric acid and sodium hydroxide. In a typical adsorption experiment, 2 mg of SBA-15 sorbent was added into 10 mL working uranium solution in a conical flask stirring at room temperature. When the adsorption equilibrium was reached, the suspension was centrifuged at 4000 rpm for 10 min. The concentration of U(VI) in the supernatant was determined by Arsenazo III Spectrophotometric Method at wavelength of 652 nm. Each experiment was repeated three times and the results given are the average values. The adsorption capacity q_t (mg g^{-1}) at time t (min) and equilibrium adsorption capacity q_e (mg g^{-1}) were calculated by the difference of the concentration between before and after adsorption according to the following equation:

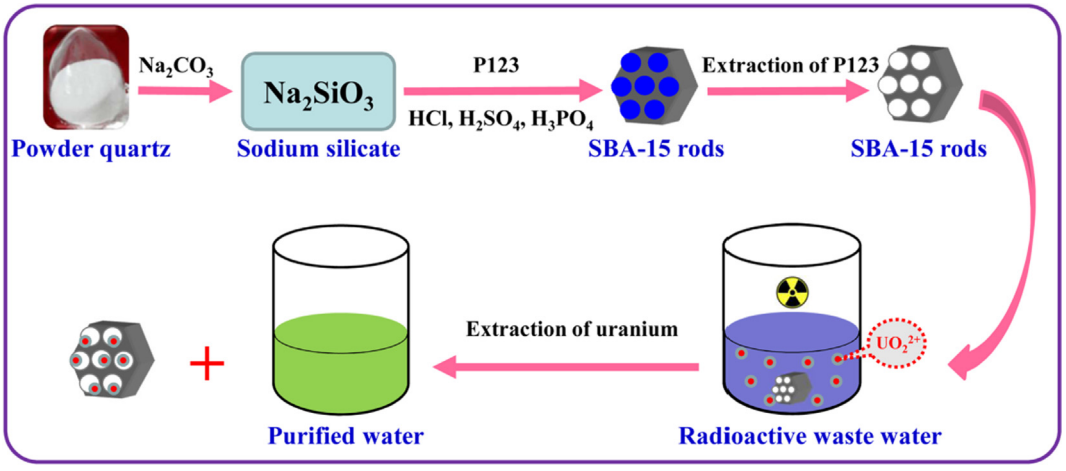

Fig. 1. The schematic diagram of research idea in this work.