



Facile synthesis of highly active hydrated yttrium oxide towards arsenate adsorption

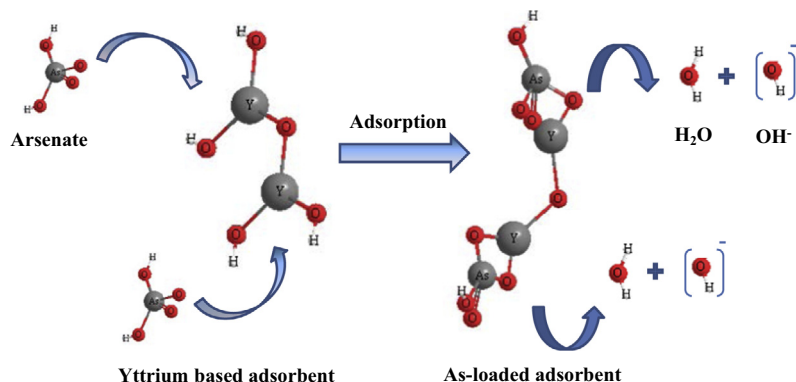


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GRAPHICAL ABSTRACT



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ABSTRACT

A novel hydrated yttrium oxide adsorbent with high capacity towards the arsenate (As(V)) adsorption was fabricated by a one-step hydrothermal process. Structure analysis identified the hydrated yttrium oxide to be Y₂O(OH)₄·1.5H₂O, which displayed as irregular rods in the range of tens to hundreds of nanometers. The adsorbent exhibited favorable As(V) adsorption efficiency in a wide pH range from 4.0 to 7.0, with the maximum adsorption capacity of 480.2 mg-As/g obtained at pH 5.0. Both the kinetics and isotherm studies demonstrated that the adsorption of the As(V) was a monolayer chemical adsorption process, in which the ion exchange between the hydroxyl groups on the hydrated yttrium oxide and arsenate anions played a key role in the uptake of the As(V). During the adsorption, the As(V) anions were replaced the hydroxyl groups and bound to the hydrated yttrium oxide via the linkage of As—O—Y. The presence of fluoride and phosphate greatly hindered the As(V) uptake on the hydrated yttrium oxide, whereas the bicarbonate, sulfate and humic acid showed insignificant impacts on the removal.

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1. Introduction

Arsenic pollution to aquatic systems has become a serious environmental issue in many regions of the world [1,2]. Generally

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speaking, arsenic is introduced into natural waters as a combined result of natural processes including weathering and mineral dissolution and anthropogenic activities such as mining, agriculture and discharge of arsenic-containing wastewaters [3,4]. The most common inorganic arsenic species in the environmental are arsenate (As(V)) and arsenite (As(III)). As(V) is usually encountered in well-oxygenated water, whereas the more toxic As(III) mainly exists in anaerobic underground water [5]. Since the As(III) is more

difficult to be removed than the As(V), As(III) is usually first oxidized to As(V), which is then treated by such technologies as adsorption.

Such technologies as co-precipitation [6], ion exchange [7,8], biotechnology [9], membrane filtration [10,11] and adsorption [12,13], have been employed for the As(V) removal. Practically speaking, the adsorption approach is more preferable than others due to its simple operation, low cost and high treatment capacity. A large amount of metal oxide adsorbents, including iron oxide [14], manganese oxide [15], zirconium oxide [16], titanium oxide [17] and complex metal oxides [18,19], have been developed for removing As(V) from wastewaters. However, the potential of these metal oxides is limited by the narrow pH range required for efficient As(V) adsorption. Therefore, developing novel adsorbents that can work in a wide pH range is of great importance for the industrial applications.

Recently, we reportedly developed a yttrium-manganese binary metal oxide adsorbent that had a good As(V) adsorption capacity in a wide range of pH [20]. It works well for removal of As(V) and As(III) in pH 4–7 and pH 4–11, respectively. The functional groups, including carbonate groups and hydroxyl groups, on the adsorbent are responsible for the excellent adsorption performance. The arsenate removal by hydrated yttrium oxide adsorbent that also contains the element of yttrium as the Y-Mn binary adsorbent has yet to be studied according to our literature studies.

In this study, a novel hydrated yttrium oxide adsorbent was first fabricated using a one-step hydrothermal method. Its adsorption behavior for the As(V) removal was investigated through a series of experimental work on adsorption kinetics, adsorption isotherm, and effects of competitive factors. The mechanism for the arsenic uptake was elucidated by several instrumental analytical studies.

2. Materials and methods

2.1. Materials

The chemicals including $Y(NO_3)_3 \cdot 6H_2O$, $NH_3 \cdot H_2O$, Na_2SO_4 , $NaHCO_3$, NaF, NaH_2PO_4 , $NaNO_3$, $Na_2HAsO_4 \cdot 7H_2O$, and humic acid sodium salt were purchased from Sigma-Aldrich. The As(V) stock solution at a concentration of 1000 mg/L was prepared by dissolving $Na_2HAsO_4 \cdot 7H_2O$ in deionized water, and was diluted by the deionized (DI) water before experiments.

2.2. Preparation of hydrated yttrium oxide adsorbent

The hydrated yttrium oxide adsorbent was prepared by a one-step hydrothermal method. A solution containing a certain amount of $Y(NO_3)_3 \cdot 6H_2O$ was magnetically stirred for 10 min, and then 25 wt.% of ammonia was dropwise added into solution under continuous stirring. After stirring for 10 min, the mixture was transferred into a 50-mL Teflon-lined autoclave at 150 °C for 3 h of hydrothermal reaction. The produced precipitate was collected by centrifugation and washed with deionized water until neutral pH was reached. Finally, the solid was dried at 60 °C overnight and grounded into fine powder to obtain the adsorbent.

2.3. Characterization of hydrated yttrium oxide adsorbent

The surface morphology of the hydrated yttrium oxide adsorbent was studied by a field emission scanning electron microscope (FESEM) (JSM6700F, JEQ, Japan). Samples were coated with a thin film of platinum to improve electric conductivity. The valence of key elements on the adsorbent surface was analyzed using an X-ray photoelectron spectroscopy (Kratos XPS system – Axis His – 165 Ultra, Shimadzu, Japan) equipped with a monochromatic

Al K α X-ray source (1486.71 eV) working at 150 W, 15 kV, and 10 mA and a base pressure of 3×10^{-8} Torr in the analytical chamber. The XPS results were collected in binding energy forms and analyzed using a non-linear least-squares curve fitting program (XPSPEAK41 Software). The C1s electron binding energy corresponding to graphitic carbon was set at 284.8 eV and used as a reference to position the other peaks. The elemental composition of adsorbent was measured by XPS spectra with the detection limits in the range of parts per thousand ranges [21,22].

The point of zero charge (pH_{PZC}) of the adsorbent was determined as previously described [23]. In brief, the adsorbent powder was suspended in 50 mL of 0.01 M $NaNO_3$ for 24 h, and then the initial pH of suspension was adjusted to a series of values between 3 and 10 with NaOH or HNO_3 . The pH value was recorded after agitating the suspension for 60 min to reach equilibrium. Next, the $NaNO_3$ was added at a concentration of 0.45 M and the final pH of suspension was measured after 3 h. The final pH value at which the ΔpH equals to zero in the ΔpH (final pH - initial pH) vs. final pH plot was determined as pH_{PZC} .

2.4. Adsorption experiments

The adsorption experiments were performed by adding 1 g of the hydrated yttrium oxide adsorbent into 100 mL of 50 mg/L As(V) solution at room temperature. Solution pH was maintained at 5.0 or 7.0 unless otherwise indicated. In the adsorption kinetics study, the samples were taken at different time intervals till the equilibrium was reached. The adsorption isotherms were measured with the As(V) concentrations ranging from 1 to 100 mg/L. The effect of solution pH on As(V) adsorption was investigated in the pH range of 3–10 by performing two sets of batch experiments. In the first experiment, the solution pH was controlled throughout the whole adsorption by adding HNO_3 or NaOH. The second experiment was conducted without pH control to observe the pH evolution during adsorption. The influence of co-existing anions and humic acid (HA) on the As(V) adsorption was studied by adding NaF, Na_2SO_4 , $NaHCO_3$, NaH_2PO_4 or HA into the As(V) solution at pH 7.0. Concentrations of NaF, Na_2SO_4 , $NaHCO_3$ and NaH_2PO_4 varied from 1 to 10 mM and the concentration of HA was in the range of 2–10 mg/L. Such concentration ranges were selected to cover the typical concentrations of corresponding ions in natural waters [24–27]. After the adsorption, the samples were taken from solution and filtered by a 0.45 μm Nylon syringe filter. The As(V) concentration was measured by an inductively coupled plasma optical emission spectrometer (ICP-OES, Thermo iCAP 6000).

3. Results and discussion

3.1. Chemical structure of adsorbent

The FESEM images demonstrate that the fabricated hydrated yttrium oxide adsorbent is composed of irregular rough rod with particle sizes ranging from tens to hundreds of nanometers (Fig. 1). The composition and chemical states of the adsorbent were examined by the XPS analysis. As shown in Fig. 2a, characteristic peaks for the yttrium and oxygen elements can be clearly found on the surface of the virgin adsorbent. The high-resolution spectrum of Y 3d shows two component peaks with the one at 158.27 eV assigned to the Y 3d $_{5/2}$ and the other one at 160.26 eV to the Y 3d $_{3/2}$ (Fig. 2b). It is therefore proposed that the yttrium in the adsorbent is presented as Y(+III). The high-resolution scan of O 1s spectrum can be divided into three component peaks, which are attributed to the yttrium oxide (Y–O), the hydroxyl group bonded with yttrium (Y–OH) and the water bounded in the adsorbent (H_2O) with the binding energies of 531.4, 531.9,

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