

A novel sort of porous ceramic foam ball with modified surface for arsenic removal from aqueous solution

Pei-sheng Liu^{*}, Guang Cui, Yi-jiao Guo, Jing-he Chen, Zi-xuan Yang
College of Nuclear Science and Technology, Beijing Normal University, Beijing 100875, China

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ABSTRACT

A sort of lightweight porous ceramic foam ball that can float on the water was recently made from the natural zeolite. The diameters of porous ball and its macroscopic pores are around 5 and 1 mm, respectively. Such ball surface could be modified to activate in different ways, i. e. loading the active alumina or desilicating the ball. Both of modified products have been investigated to adsorb the toxic ion of arsenic (As) in water effectively. This kind of porous ceramic ball was prepared by powder sintering, using natural clinoptilolite powder as the primary raw material. An active-alumina-loaded porous ceramic product was generated by sol-gel method with pseudoboehmite (AlOOH) as the main composition. The active Al atom on the porous ball surface could also be achieved by desilicating the ceramic ball in the NaOH solution. Such a desilicated product exhibited a good activity to adsorb the arsenic ion although carrying no active alumina. With this effective porous structure, the present products can be expected to become practically advantageous.

1. Introduction

Toxic ions in water would cause a notable environmental problem and bring a serious threat to the human health^[1–3]. Arsenic (As) is an element widely existing in the nature, which can increase the incidence of cancerization of skin, lung, bladder, liver and kidney^[2,3]. In water, As may result mainly from the permeability of As-contained rocks or sediments, and the use of As-contained pesticides or wood-preserved^[2–4]. In the surface water containing oxygen, As(V) is the dominant arsenic form; mostly H_2AsO_4^- when $\text{pH} < 7$ and HAsO_4^{2-} when $\text{pH} > 7$ ^[5].

In addition to the arsenic species in the natural water, the relevant industries will generate a relatively large amount of arsenic ions in the production process. When these arsenic ions get into the groundwater circulation system, they will seriously affect the safety of drinking water. Especially in the less developed regions, the arsenic ion pollution is more direct and inevitable.

The treatment of As-contained wastewater has become a very important work which is difficult to deal with for environmental protection. The commonly used methods for removing As include the coagula-

tion sedimentation, ion exchange, adsorption, biological treatment, electrocoagulation, and membrane separation, etc.^[2,3,6–8]. These methods may be effective in some cases, but will also bring some unwelcome results. For instance, the sedimentation technique is relatively perfect and extensively used, but it will generate a lot of waste residue to cause the second pollution. The biological treatment need be very strict, and the membrane separation process is quite expensive. Comparatively, the adsorption method, which uses the active surface of adsorbent to achieve the goal of As removal, is simple and easy to operate, and exhibits a promising prospect in practical applications^[3,5,8]. This technique, however, will lead to the second pollution due to using powdered adsorbents.

In virtue of the simple operation, good As-removal effect and large treatment capacity, the active alumina adsorption technique has attracted a lot of attention^[7,9]. Besides, other adsorbent species have been found to also yield a quite good effect of As adsorption, such as iron oxide^[10]. Active alumina balls are usually adopted to treat the As-polluted water in case of emergency^[7,9]. In practice, the solid alumina balls cannot be favorable to the diffusion of pollu-

^{*} Corresponding author. Prof., Ph.D.
E-mail address: Liu996@263.net (P. S. Liu).

tants in water, and the inner active alumina cannot be well utilized, while the hollow alumina balls commercially available are easy to collapse during the service. Thereby, the combination of active alumina and the porous ball should be considered. Porous ceramic foam products may be advantageous to a practical application in the polluted water treatment^[11–13], but they can only have a higher density than water. Resultantly, a novel sort of lightweight porous ceramic foam ball carrying active alumina was made to simultaneously solve the above-mentioned problems. This kind of loaded ball may float on water, or suspend or sink in water, so as to be conducive to the water treatment process. The adsorption function of active alumina resulted from the active aluminum (Al) atom, so desilicating the porous ceramic ball was also developed to obtain a great deal of exposed active Al atoms.

2. Experimental

2.1. Preparation of porous ceramic balls

Using the natural zeolite powder as the primary raw material, the present porous ceramic ball was prepared by powder sintering (Fig. 1). Firstly, the zeolite powder (natural clinoptilolite, $\text{Na}_8[\text{Al}_8\text{Si}_{40}\text{O}_{96}] \cdot 39\text{H}_2\text{O}$), polymer ball and binder were uniformly blended with the addition of some assistant species, and the green zeolite-base composite product was obtained after stirring of a ball-forming machine. This green product was dried and then put inside a muffle furnace to heat from room temperature up to 1000 °C to sinter for 1 h. After sintering, the finished product presented a novel sort of porous ceramic foam ball.

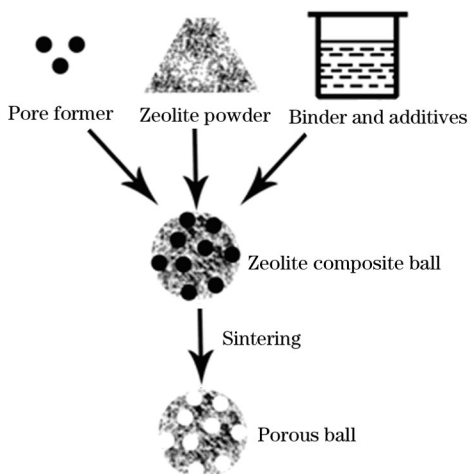


Fig. 1. Process for preparing porous ceramic ball.

2.2. Loading active alumina on porous ceramic ball surface

A layer of active alumina was loaded on the above-mentioned sintered porous ball by sol-gel method, with pseudoboehmite as the main composition of the

sol and gel. The sol was prepared by the nitric acid method, and the white gel formed based on the proportioning: $m(\text{AlOOH}) : V(\text{H}_2\text{O}) : V(\text{HNO}_3) = 1 \text{ g} : 10.0 \text{ mL} : 0.4 \text{ mL}$. The porous balls were placed in deionized water, and ultrasonically cleaned for 30 min to remove the impurity on the surface. After drying, these balls were immersed in the pseudoboehmite sol to experience an ultrasonic vibration for 60 min, and then taken out to age in air at room temperature for 12 h. With this process, the sol loaded on the surface of the porous ball could well enter the inside pores. The sol-loaded porous balls afterwards went through a heat treatment to transform the sol into $\gamma\text{-Al}_2\text{O}_3$, an active alumina.

2.3. Desilication of porous ceramic ball

The desilication was carried out as follows: the porous ball was firstly put in the 1 mol/L NaOH solution at 60 °C for 10 h, and then was flushed with deionized water until pH became neutral. After drying at 100 °C, a desilicated product yielded.

2.4. As/Cs adsorption experiment

The aqueous solution of $\text{NaH}_2\text{AsO}_4 \cdot 7\text{H}_2\text{O}$ (1 mg/L) was used in the As adsorption experiment, with the pH value of 6.4. An amount of 2 g of the active alumina loaded porous ball or the desilicated ball product was put in the arsenic solution of 1 L to test the As adsorption.

Based on the potential awareness to deal with those like Fukushima accident of Japan in 2011, the experiment was also tried to adsorb Cs^+ in water by the present desilicated porous ball. In this experiment, the porous ball (2 g) was put in a volume of 50 mL of the 100×10^{-6} CsCl solution at pH=7.

2.5. Characterization

The sintered porous ceramic product and the alumina loaded on the porous ball were identified by X-ray diffraction (XRD) with $\text{CuK}\alpha$ radiation, using a X'Pert PRO MPD diffractometer, and the surface morphologies of the product were examined by scanning electron microscopy (SEM), using a Hitachi SEM 4800 device. The arsenic concentration in the NaH_2AsO_4 solution was determined by atomic fluorescence spectrometry (AFS), using a SPECTRO ARCOS ICP-OES spectrometer, and the adsorption efficiency (absorbance, η) was calculated by:

$$\eta = [(C_0 - C)/C_0] \times 100\% \quad (1)$$

where, C_0 is the initial arsenic concentration; and C is the timely arsenic concentration in the tested solution.

3. Results and Discussion

3.1. Sintered porous ceramic ball

The resultant sintered porous ball is around 5 mm

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