

# Characteristics of molybdate-impregnated chitosan beads (MICB) in terms of arsenic removal from water and the application of a MICB-packed column to remove arsenic from wastewater

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## Abstract

The removal of arsenic (As) species, such as As(III) and As(V), from water by molybdate-impregnated chitosan beads (MICB) in both batch and continuous operations was studied. The effects of pH, temperature, coexisting ions, and arsenic concentrations were studied in batch tests. Studies on the kinetic adsorption of MICB, the recovery of arsenic by the desorption solution, and the reuse of MICB were also carried out. The practicality and efficiency of an MICB-packed column on arsenic removal were evaluated in a continuous system on industrial arsenic-containing wastewater discharged during the manufacture of GaAs supports. The results indicate that MICB favor the adsorption of both As(V) and As(III). The optimal pH value for As(III) and As(V) removal was 5. The adsorption of arsenic on the MICB is most likely an exothermic reaction. The effect of coexisting ions was varied and depended on their concentrations and species. The optimal desorption solution for arsenic recovery was 1 M H<sub>2</sub>SO<sub>4</sub>, which resulted in a 95% efficiency for As(III) and 99% for As(V). In the continuous tests, the MICB-packed column exhibited excellent arsenic removal from wastewater without any pretreatment. These results provide strong evidence of the potential of MICB for removing As from industrial wastewaters.

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## 1. Introduction

The presence of arsenic in groundwater as the result of anthropogenic pollution has become a major public health problem concern in many countries, such as India, Vietnam, Bangladesh, Taiwan, China, USA, Argentina, Chile, Poland, Canada, Hungary, Japan, and Mexico (Pokhrel and Viraraghavan, 2006; Dinesh et al., 2007). An additional problem facing many countries, such as Taiwan, India, Bangladesh, and Vietnam, among others, is the presence of naturally occurring high levels of arsenic in the

groundwater (Pokhrel and Viraraghavan, 2006). Arsenic contamination of groundwater is problematic throughout the world because most people rely on tube wells as a source of drinking water. About 40–60% of the people in Bangladesh have been estimated to be adversely affected by arsenic-contaminated drinking water (Jang et al., 2006). Thus, the removal of arsenic from industrial effluents, groundwater, and even drinking water systems has become a very important issue. The World Health Organization (WHO) recommends a maximum admissible concentration of arsenic of 10 µg l<sup>-1</sup> in drinking water (WHO, 1993). In Taiwan, the standard allowances of arsenic in industrial effluents and drinking water are set at 500 µg l<sup>-1</sup> and 10 µg l<sup>-1</sup>, respectively (Chen and Chung, 2006), with most of the arsenic compounds found in the

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former deriving from the by-products of pesticide, chemical, pharmaceutical, and semiconductor manufacturing processes (Tseng et al., 2005).

With the advances in science and technology, the focus on arsenic pollution has shifted from treating groundwater to treating industrial wastewater; this is particularly true for the effluents from the manufacturing of GaAs supports in a number of Asian areas. These effluents contain relatively high As(V) and trace As(III) concentrations compared to groundwater polluted by arsenic compounds. In addition, these wastewaters from electronics manufacturing processes are characterized by fewer variations of chemical species in the wastewater relative to those found in the groundwater.

Many arsenic removal technologies have been developed. These technologies are generally classified into three types in accordance with the removal system – precipitation, membrane, and adsorption – but combinations of these processes or technologies can be used if required. Aluminum salt and iron salt are useful coagulants in removing arsenic compounds by the coagulation precipitation method (Han et al., 2002). However, the production of highly toxic sludge has limited the application of such systems (Dinesh and Charles, 2007). Arsenic in natural waters is mostly found in inorganic form as oxyanions of trivalent arsenite As(III) or as pentavalent arsenate As(V). Membrane technology is more effective at removing As(V) than As(III) (Ning, 2002; Xu et al., 2002). Therefore, the addition of an extra oxidizing agent (i.e. chlorine) to the water being treated is usually required in order to convert As(III) to As(V). Unfortunately, these agents often injure the membranes, substantially shortening their life time. The adsorption technique is regarded as a promising method or process because it has been successfully tested in many different systems (Xu et al., 2002). Activated alumina, natural iron-enriched samples, polymeric hybrid, natural solids, waste tea fungal, iron oxide-loaded slag, and chromate copper arsenate (CCA)-treated wood have all been applied to the removal of arsenic from water (Elizalde-González et al., 2001; Demarco et al., 2003; Singh and Pant, 2004; Deschamps et al., 2005; Kartal and Imamura, 2005; Zhang and Itoh, 2005; Murugesana et al., 2006). In such systems, an efficient, cheap, and environmentally friendly adsorbent is very important.

Chitosan is transformed polysaccharide obtained by the de-*N*-acetylation of chitin, which makes up the shells of crustaceans such as crabs and shrimps (Jang et al., 2004). It is biodegradable, biocompatible, and nontoxic, making it an environmentally friendly substance (Kurita, 1998; Chen et al., 2007). The free amino groups of chitosan can serve as sites of coordination binding for metals, and some studies have demonstrated chitosan's adsorption capacity for metal ions such as Cd(II), Cu(II), Cr(III), Pb(II), Zn(II), Ni(II), Hg(II), As(III), and As(V) (Onsoy and Skaugrud, 1990). Furthermore, some chemical modifications of chitosan have also been carried out to introduce novel functions into this biopolymer (Chen et al., 2007).

For example, chitosan impregnated with molybdate, in the form of “molybdate-impregnated chitosan beads” (MICB) has been used to improve the absorption of metals and organic contaminants (Chen and Chung, 2007). Chitosan has a very strong affinity for molybdate, and molybdate ions are known for their ability to form complexes with metal ions.

Although some studies have used MICB as a sorbent to remove arsenic from water, these studies focused on the arsenic sorption mechanism, were evaluated under limited operating conditions, and suffered from a lack of continuous and actual environmental data (Dambies et al., 2000, 2002). Hence, in the study reported here, we investigated arsenic removal using MICB in both batch and continuous experiments and evaluated a number of important factors affecting arsenic adsorption by MICB. We also tested the applicability of a MICB-packed column to the removal of arsenic from wastewater discharged during the manufacturing of GaAs supports. The overall objective of our study was to develop a MICB-packed column that enhances the removal of arsenic from actual wastewater containing high concentrations of arsenic compounds in a one-step operation without any pretreatment steps.

## 2. Methods

### 2.1. Materials

Chitosan, derived from shrimp shells, with a deacetylation degree of 90% was obtained from the Application Chemistry Company (Kaohsiung, Taiwan). Its characteristics were  $pK_a = 5.6$ , and weight average molecular mass ( $MW_w$ ) =  $1.5 \times 10^6$  g mol<sup>-1</sup>. Stock solutions of As(III) and As(V) were prepared by dissolving analytical grade reagents NaAsO<sub>2</sub> and Na<sub>2</sub>HAsO<sub>4</sub> · 7H<sub>2</sub>O in deionized water. NaAsO<sub>2</sub> and Na<sub>2</sub>HAsO<sub>4</sub> · 7H<sub>2</sub>O were purchased from Merck. All other chemicals were purchased from Sigma Chemical Co. unless otherwise specified.

### 2.2. Preparation of MICB

Powdered chitosan bead was dissolved in 1 M acetic acid to give a final concentration of 1% (w v<sup>-1</sup>), and the solution was dropped to 1 M NaOH solution by microsyringe. The reaction solution was stirred gently for 16 h at room temperature, following which the chitosan beads were washed three times by deionized water. The wet chitosan beads were subsequently brought into contact with 5 g Mo l<sup>-1</sup> of ammonium heptamolybdate solution at pH 3. The reaction solution was stirred gently for 24 h at room temperature to form the well structure of the molybdate-impregnated chitosan bead, and the MICB was dried at 60 °C for reuse. The water content, average size, and molybdate content of these beads were about 94.6%, 2.5 mm, and 650 mg Mo g-dry MICB<sup>-1</sup>, respectively.

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