



# Concurrent removal of cadmium and benzene from aqueous solution by powdered activated carbon impregnated alginate beads



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## ABSTRACT

Applicability of a powdered activated carbon impregnated alginate beads (ABPAC) was examined toward simultaneous removal of heavy metal (cadmium) and volatile organic compound (benzene) from water. Sorption of cadmium and benzene by ABPAC was investigated in both single and binary systems as a function of contact time (1 h–11 d), initial pH (4–7), and initial concentration of cadmium and benzene (10–600 mg L<sup>-1</sup>). The sorption capacity of APBAC for cadmium and benzene was 149.32 and 89.60 mg g<sup>-1</sup>, respectively. Sorption kinetics was well described by the pseudo-second-order model with correlation coefficients, R<sup>2</sup> > 0.994. Cadmium sorption capacity increased from 41.76 to 56.09 mg g<sup>-1</sup> with increasing pH from 4 to 7, while benzene sorption was not much influenced. The Langmuir model best correlated with adsorption of cadmium and benzene on ABPAC. Our results indicated no competition effects in a binary-contaminant system, which proves the practical utility of ABPAC for the concurrent sorptive removal of aqueous phase heavy metals and volatile organic compounds.

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## 1. Introduction

In recent years, industrialization has led to increased disposal of inorganic and organic contaminants into the environment. Soil and groundwater in the vicinity of certain industrial estates, landfills, and dump sites of chemical substances have been contaminated with heavy metals and volatile organic compounds (VOCs), exceeding the ground water standards (Pollution Control Dept., 2012; Reddy, 2008). Contamination of soil and water by heavy metals and VOCs poses a threat to human beings as they are potentially toxic and carcinogenic (Aksu and Gönen, 2006). Cadmium imposes lethal effects on bones, kidney, liver, and other vital organs (Maine et al., 2001), while benzene causes structural and numerical chromosomal aberrations in humans (Wilbur et al., 2007). The World Health Organization has mandated the maximum level for cadmium and benzene in drinking water at 3 and 10 ppb, respectively (WHO, 2011). Considerable efforts have been made to remove heavy metals and VOCs from contaminated waters (Farhadian et al., 2008; Fu and Wang, 2011) without noticeable study

on the simultaneous removal of both heavy metals and organic compounds (Chao et al., 2013).

Adsorption has been recognized as an effective method for purification of water containing low levels of less-biodegradable, dissolved toxins including heavy metals and VOCs (Demirbas, 2008). Among many commercial adsorbents, activated carbon (AC) is most widely used adsorbent in water/wastewater purification processes (Cho et al., 2011) due to its affinity for a wide spectrum of organic contaminants. However, AC has a relatively low affinity for heavy metals. Composite adsorbents, developed by immobilization of carbonaceous materials and adsorbents possessing a high affinity for metals into a support matrix, have found application in the removal of heavy metals or VOCs from single-contaminant systems (Jain et al., 2013; Lin et al., 2005; Sui et al., 2012). Composite adsorbents synthesized by entrapment of the powdered adsorbent in porous matrix show comparable removal efficiency than the pristine (crystalline and granular) adsorbents, as they contain nano- or micro-sized adsorbents which are highly reactive with high surface area (Khan et al., 2013; Lazaridis et al., 2007; Lin et al., 2005).

Different polymers such as alginate, polyvinyl alcohol, chitin/chitosan, cellulose, and polyacrylamide can be used to immobilize powdered adsorbents (Rocher et al., 2008). Among these, alginate, an anionic polysaccharide extracted from brown algae, has been extensively

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investigated as it is non-toxic, biocompatible, biodegradable, and highly available (Lazaridis et al., 2007; Mata et al., 2009). A carboxyl and other oxygen-containing functional groups on alginate uptake metal ions under moderate to alkaline pH conditions, making it a good adsorbent for removing heavy metals (Davis et al., 2003).

Treatment of water streams containing dissimilar binary contaminants such as cadmium and benzene is difficult due to sorption competition or inhibition. Composite adsorbents might remove both benzene and cadmium due to the presence of multiple potential sites on their surfaces; however, they have not yet been extensively investigated for mixed contaminants. In this regard, a composite adsorbent was developed by immobilizing commercial powdered activated carbon (PAC) in calcium-alginate beads and was evaluated for the simultaneous removal of cadmium and benzene in a binary system. Sorption kinetics and isotherm experiments in the single and binary-contaminant system using powdered activated carbon impregnated alginate beads (ABPAC) were performed, and the results were compared with those obtained using pure alginate beads (AB) as well as PAC. In addition, pseudo-second-order kinetic and Langmuir and Freundlich isotherm parameters were also employed in this study.

## 2. Materials and methods

### 2.1. Chemicals

All the chemicals used in this study were analytical grade. Sodium alginate (medium viscosity) and benzene (>99% purity) were obtained from Sigma–Aldrich Co., USA; cadmium nitrate was obtained from Junsei Chemical Co., Japan; calcium chloride was obtained from Tedia Co., USA. The stock standard solution of cadmium was prepared by dissolving desired amount of the nitrate salt in deionized (DI) water.

### 2.2. Preparation of powdered activated carbon impregnated alginate beads

Sodium alginate (3%, w/V) was mixed with 400 mL of DI water at 60 °C under stirring condition. Commercial powdered activated carbon (PAC; 3%, w/V) was then poured into the alginate solution and mixed with a magnetic stirrer for 12 h to obtain a homogeneous mixture of the PAC and alginate. Water insoluble composite beads were synthesized by introducing the gel matrix into 3% CaCl<sub>2</sub> solution through a 16 gauge (1.7 mm) needle. The produced beads were left in the CaCl<sub>2</sub> solution for 12 h for complete gelation and were washed five times with DI water to remove excess calcium from the beads. The beads were then dried at room temperature for 1 day. After drying, the size of the beads was reduced by two-thirds with a diameter of approximately 1.0 mm. Beads without impregnated carbon were also produced by the same method for comparison purpose.

### 2.3. Adsorption experiment

Batch adsorption experiments were performed at 25 °C in 50 mL glass vials by adding 0.04 g (1.0 g L<sup>-1</sup>) of adsorbent to the experimental solution and the vials were sealed immediately to prevent volatilization. Sorption studies on heavy metals and organics including kinetics and isotherm experiments have been mostly investigated at an adsorbent dose between 0.5 and 2.0 g L<sup>-1</sup> (Mata et al., 2009; González et al., 2015). Control experiments (without adsorbent) were also performed to determine the loss of adsorbate due to adsorption to vial walls or volatilization during preparation and sampling. The initial solution pH (without pH adjustment) was in the range of 5–5.5. Each experiment was performed in triplicate and average values were reported with standard deviation.

Kinetics experiments were conducted in 50 mL screw cap glass vials containing 40 mL of 100 mg L<sup>-1</sup> of benzene or cadmium, and 0.04 g (1.0 g L<sup>-1</sup>) of adsorbent. The vials were mixed on a temperature-controlled rotary shaker and samples were withdrawn at predefined

time intervals. Equilibrium sorption experiments for cadmium and benzene were performed by varying initial concentration ranging from 10 to 600 mg L<sup>-1</sup>. In the binary-contaminant system, both sorbates were added in equal amounts, such that the total sorbate concentration was doubled compared to the single-contaminant system. The influence of one contaminant on the sorption of another contaminant was evaluated through direct comparison of kinetics and equilibrium of cadmium and benzene sorption in binary-contaminant system with that in the single system. The adsorption capacity  $q_e$  (mg g<sup>-1</sup>) of the adsorbent was calculated using Eq. (1):

$$q_e = \frac{(C_0 - C_e)V}{m} \quad (1)$$

where  $C_0$  (mg L<sup>-1</sup>) is the initial concentration,  $C_e$  (mg L<sup>-1</sup>) is the equilibrium concentration remaining in the solution,  $V$  (L) is the volume of the solution, and  $m$  (g) is weight of the adsorbent. The effect of pH on cadmium and benzene removal by ABPAC in single and binary-contaminant systems was also studied. An equilibrium sorption on ABPAC was investigated for initial benzene and/or cadmium concentration of 100 mg L<sup>-1</sup> with the initial solution pH being adjusted to 4–7.

Analyses of sorption kinetics and isotherm data are important for determining how the sorption process occurs; this helps to determine the nature of sorption process and to evaluate the maximum sorption capacity of the adsorbent (Iqbal et al., 2009). Kinetics data were fitted to pseudo-second-order model and isotherm data were fitted to Langmuir and Freundlich isotherm models.

### 2.4. Analytical methods

The BET surface area of PAC, ABPAC, and AB was determined by the N<sub>2</sub> adsorption/desorption method at 77 K using an ASAP 2010 (Micromeritics, USA). Gas chromatography (DS6200, Donam Instruments Inc., Korea) equipped with flame ionization detector (GC-FID) and inductively coupled plasma optical emission spectroscopy (ICP-OES, 3000 DV, Thermo Jarrel Ash Corp., USA) were used to measure the concentration of benzene and cadmium, respectively. Sample preparation and injection into GC were done by the static headspace method described in our earlier study (Khan et al., 2010). A DB-624 column (Agilent, USA) with a length of 30 m and an internal diameter of 0.53 mm was used. Nitrogen was used as a carrier gas and the GC-FID was operated at an injection temperature of 250 °C, detection temperature of 270 °C, and oven temperature of 70 °C. ICP-OES was equipped with dual detector (CCD, UV-Vis), radiofrequency source (40 MHz), temperature control (10,000–5600 K) and WinLab32™ ICP continuous-Diagnostics software. Liquid argon (≥99.996%) and nitrogen (≥99.996%) were used as torch and purge gases, respectively. More information about the standard operating conditions for ICP-OES instrument is available as supporting information (Table S1).

## 3. Results and discussion

### 3.1. Characterization of adsorbents

BET surface areas of PAC, ABPAC, and AB were 1023.11, 488.79, and 2.37 m<sup>2</sup> g<sup>-1</sup>, respectively. Lower surface area of composite adsorbent than powdered adsorbent could be due to agglomeration of particles, blocking of particles' pore by support matrix and lower surface area of the support than powdered adsorbent. The surface area of beads after impregnation of PAC was high, which indicated that the composite adsorbent was highly porous. FTIR analysis was performed for the adsorbents (PAC and ABPAC) before and after sorption studies, but no significant changes were observed (supporting information, Fig. S1).

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