



## Evaluation of metal oxides and activated carbon for lead removal: Kinetics, isotherms, column tests, and the role of co-existing ions

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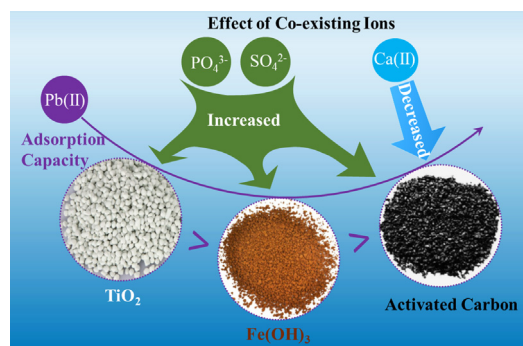
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### HIGHLIGHTS

- Metal oxides showed higher Pb(II) removal ability than AC.
- Ca(II) reduced Pb(II) removal ability of AC, but had no effect on metal oxides.
- Both Ca(II) and Pb(II) were adsorbed on AC as inner-sphere surface complexes.
- Orthophosphate enhanced Pb(II) removal by synergistic adsorption.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 25 June 2018

Received in revised form 1 August 2018

Accepted 2 August 2018

Available online 03 August 2018

Editor: Jay Gan

#### Keywords:

Lead

Adsorption

Activated carbon

Metal oxides

Co-existing ions

### ABSTRACT

Activated carbon (AC) is commonly used in faucet and pitcher filters for lead (Pb(II)) removal in homes. This study evaluated the Pb(II) removal performance of AC and metal oxides (e.g. Fe(OH)<sub>3</sub> and TiO<sub>2</sub>), as well as the co-existing ions' effect on Pb(II) removal. Results showed that metal oxides had higher adsorption capacity (28.9–51.5 mg/g) than AC (21.2 mg/g). Pb(II) was inner-spherically adsorbed onto both AC and metal oxides surfaces. Among various metal ions, calcium (Ca(II)) demonstrated dramatic effects on Pb(II) removal ability of AC, while it had no effect on Pb(II) adsorption by metal oxides. This difference resulted from the inner- and outer-sphere adsorption of Ca(II) on AC and metal oxides, respectively. The presence of orthophosphate (orth-P) and sulfate enhanced Pb(II) removal by those three adsorbents, whereas carbonate and silicate had negligible effect on Pb(II) adsorption. Interestingly, while the orth-P was usually used as corrosion inhibitor because of the formation of lead-phosphate coprecipitate, we found that the enhanced effect of orth-P on Pb(II) removal was mainly due to the synergistic adsorption. This study provides valuable information for the selection of effective adsorbents for Pb(II) removal and is helpful for understanding the roles of co-existing ions on it.

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## 1. Introduction

Recently, the drinking water crisis in Flint, Michigan, United States (U.S.), which results from high lead (Pb(II)) concentrations in drinking water, has attracted a lot of attention (Torrice, 2016). Lead poisoning

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crises have also occurred in Washington, D.C. (Edwards et al., 2009), Pennsylvania (Frostenson, 2016; Mankikar et al., 2016) and Virginia (Pieper et al., 2015), suggesting that it is still a serious and nationwide drinking water contamination and human health problem. High levels of Pb(II) in the drinking water are caused by corrosion of the old lead-containing plumbing systems (i.e., lead solder and leaded brass) (Xie and Giammar, 2011). The drinking water standards for Pb(II) set by the U.S. Environmental Protection Agency (USEPA) and World Health Organization (WHO) are 15 and 10  $\mu\text{g/L}$ , respectively. Exposure to Pb(II) beyond these guidelines may cause health problems such as abortion, stillbirths, sterility, neo-natal deaths and intellectual disability (Edwards et al., 2009; Nordberg et al., 2014).

Point-of-Use (POU) devices are widely used for tap water purification in the U.S. Numerous POU devices have been certified for Pb(II) removal by the National Sanitation Foundation (NSF) International. Over 200,000 of the certified POU devices were distributed for removal of Pb(II) in Flint in 2016. However, it was reported that those POU devices had low efficiency for Pb(II) removal, especially for water with high Pb(II) concentrations (Deshommes et al., 2010). Therefore, it is urgent to identify and develop more efficient POU devices for Pb(II) removal.

Activated carbon (AC) is usually used as an adsorbent in POU devices. On the other hand, numerous studies have indicated high performance for Pb(II) removal using metal oxides (Ali, 2012; Naiya et al., 2009; Ponder et al., 2000; Recillas et al., 2011; Vohra and Davis, 1997). In a study regarding selection of POU adsorbents for Pb(II) removal, AC coated with synthetic zeolite, natural  $\text{MnO}_2$ , and natural zeolite performed better than 8 other adsorbents (Sublet et al., 2003). Moreover, to improve the Pb(II) removal ability, metal oxides modified carbon materials and other hybrid materials have been widely studied (Table S1). Nevertheless, the current commonly used commercial metal oxides such as  $\text{Fe}(\text{OH})_3$  and  $\text{TiO}_2$  have not been compared with AC in terms of Pb(II) removal efficiency. In addition, although Pb(II) removal using AC (Chen and Wang, 2000; Imamoglu and Tekir, 2008; Jusoh et al., 2007; Momčilović et al., 2011; Monser and Adhoum, 2002; Netzer and Hughes, 1984; Wang et al., 2010) has been broadly studied for a long time, little study has been reported regarding the effect of co-existing ions on Pb(II) removal by AC.

The objectives of this study are to compare the Pb(II) removal ability of AC and metal oxides ( $\text{Fe}(\text{OH})_3$  and  $\text{TiO}_2$ ), and evaluate the effect of co-existing ions on Pb(II) adsorption. Batch experiments, including kinetics, isotherms, and pH adsorption edge, were employed to determine Pb(II) removal ability of AC,  $\text{Fe}(\text{OH})_3$ , and  $\text{TiO}_2$ . Meanwhile, the effects of common co-existing ions on the Pb(II) removal ability of those adsorbents were also investigated. The point of zero charge (PZC) of these three adsorbents was measured to explore the interaction mechanisms between Pb(II) or the co-existing ions and the adsorbents. Filtration tests using Pb(II)-spiked tap water were conducted to evaluate the performance of the adsorbents.

## 2. Materials and methods

### 2.1. Materials

Stock solutions of 1000 mg/L Pb(II), calcium ( $\text{Ca}(\text{II})$ ), orthophosphate (orth-P), sulfate ( $\text{SO}_4^{2-}$ ), carbonate ( $\text{CO}_3^{2-}$ ), and silicate ( $\text{SiO}_3^{2-}$ ) were prepared by dissolving  $\text{PbCl}_2$ ,  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ ,  $\text{NaH}_2\text{PO}_4$ ,  $\text{Na}_2\text{SO}_4$ ,  $\text{Na}_2\text{CO}_3$ , and  $\text{Na}_2\text{SiO}_3$  in deionized (DI) water (18.2 M $\Omega$ , Milli-Q), respectively. All chemicals used were of analytical grade, and purchased from Fisher or Sigma-Aldrich, U.S. Five AC adsorbents were used in the experiments, including a virgin AC (Nichem Company, U.S.) and four AC samples taken from Brita (BR), Pelican (PE), Pura (PU), and ZeroWater (ZE) POU filter cartridges.  $\text{Fe}(\text{OH})_3$ ,  $\text{TiO}_2$ , and activated alumina (AA) were commercially available and obtained from AdEdge Water Technologies (U.S.), Graver Technologies (U.S.), and Alcan (Canada), respectively. All the adsorbents were sieved to 30–50 mesh (0.3–0.6 mm) for the experiments.

### 2.2. Batch experiments

Batch adsorption experiments including kinetics, isotherms, and pH adsorption edge were conducted to compare the efficiencies of the adsorbents. In a typical adsorption experiment, adsorbents were mixed with Pb(II) solution at the desired pH values. The samples were mixed on a rotator at 40 rpm for 4 h, filtered through a 0.2  $\mu\text{m}$  microfilter (Fisher, U.S.), and then acidified to  $\text{pH} < 1$  using  $\text{HNO}_3$ . The Pb(II) concentration was determined by graphite furnace atomic absorption spectrometry (GFAAS, Agilent Z240, U.S.).

The kinetics of Pb(II) adsorption on 0.4 and 0.1 g/L virgin AC,  $\text{Fe}(\text{OH})_3$ , and  $\text{TiO}_2$  were studied in 1 L 0.01 mol/L NaCl solutions with 10 and 1 mg/L Pb(II), respectively. In the isotherm study, the adsorbent concentration was 0.4 g/L, and the Pb(II) concentration was varied in the range from 0.1 to 25 mg/L. In the kinetic and isotherm studies, solution pH was controlled at  $5.0 \pm 0.2$  using NaOH and HCl. The pH adsorption edge experiments of 10 mg/L Pb(II) on 0.4 g/L adsorbents and 1 mg/L Pb(II) on 0.1 g/L adsorbents were conducted as a function of solution pH.

To explore the mechanism of the increase in Pb(II) removal in the presence of orth-P, orth-P (1 mg/L) removal by different adsorbents with and without 1 mg/L Pb(II) was studied as a function of pH. In a control test, precipitate formation was explored by measuring orth-P in 0.01 M NaCl solutions with 1 mg/L Pb(II) at different pH in the absence of adsorbents.

In another isotherm experiment,  $\text{Fe}(\text{OH})_3$ ,  $\text{TiO}_2$ , and the AC used in the above experiments, as well as four other types of commercial AC mentioned above, were added into Pb(II)-spiked aged tap water to compare their Pb(II) removal ability in real water. The pH was controlled at neutral ( $7.0 \pm 0.2$ ).

To explore the reusability of those adsorbents for Pb(II) removal, 1 g/L  $\text{Fe}(\text{OH})_3$ ,  $\text{TiO}_2$ , and AC were separately mixed with 10 mg/L Pb(II) and 0.01 mol/L NaCl. After 4 h mixing, the suspension was separated, and the residual adsorbents were regenerated using 50 mL 5 mM  $\text{HNO}_3$ . Moreover, the desorbed Pb(II) in solution was tested at 10, 20, and 30 min to investigate the optimum desorption mixing time. After regeneration, the adsorbents were washed by 20 mL DI water for 3 times for the next re-use cycle.

### 2.3. Column filtration tests

Four columns with the internal diameter of 1.5 cm were filled with 20 mL of AC, AA,  $\text{Fe}(\text{OH})_3$ , and  $\text{TiO}_2$ , corresponding to the masses of 10.08, 15.62, 10.02, and 14.57 g, respectively. The tap water was firstly exposed to the air in a 50 L tank for at least 8 h, and then the Pb(II) was added into the tank. After 1 h mixing, the Pb(II)-spiked tap water was siphoned to another 150 L tank, and then passed through the columns in a downward flow mode with a flow rate of 10 mL/min. The Pb(II) spiked tap water was prepared twice per day, and the compositions of the tap water included 16–25 mg/L  $\text{Ca}(\text{II})$ , 43–53 mg/L sodium, 87–103 mg/L chloride, and 0–12 mg/L sulfate. Samples of effluent were collected and acidified with  $\text{HNO}_3$  for GFAAS analysis.

### 2.4. Zeta ( $\zeta$ ) potential measurements

AC and  $\text{Fe}(\text{OH})_3$  powders (>200 mesh) were employed for measuring the  $\zeta$  potential by a Zetasizer Nano ZS (Malvern Instrument Ltd., UK). The pH of the suspension containing 0.01 g/L of adsorbent in 0.01 mol/L NaCl was adjusted to the designated pH range using 0.1 mol/L NaOH and HCl solutions. The suspension samples were mixed on a rotator for 24 h and the final pH was measured. The reported  $\zeta$  potential values were the average of three measurements. These experiments were performed under  $\text{N}_2$  atmosphere to eliminate  $\text{CO}_2$  from the system.

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