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## CONTENTS

**Aquatic environment**

- Comparison of conventional and inverted A<sup>2</sup>/O processes: Phosphorus release and uptake behaviors  
Rong Qi, Tao Yu, Zheng Li, Dong Li, Takashi Mino, Tadashi Shoji, Kochi Fujie, Min Yang ..... 571
- Distribution of heavy metals in sediments of the Pearl River Estuary, Southern China: Implications for sources and historical changes  
Feng Ye, Xiaoping Huang, Dawen Zhang, Lei Tian, Yanyi Zeng ..... 579
- Removal of arsenate and arsenite from aqueous solution by waste cast iron  
Nag-Choul Choi, Song-Bae Kim, Soon-Oh Kim, Jae-Won Lee, Jun-Boum Park ..... 589
- Effect of artificial aeration on the performance of vertical-flow constructed wetland treating heavily polluted river water  
Huiyu Dong, Zhimin Qiang, Tinggang Li, Hui Jin, Weidong Chen ..... 596
- A 60-year sedimentary record of natural and anthropogenic impacts on Lake Chenghai, China  
Fengyu Zan, Shouliang Huo, Beidou Xi, Jingtian Zhang, Haiqing Liao, Yue Wang, Kevin M. Yeager ..... 602
- Preparation and application of amino functionalized mesoporous nanofiber membrane via electrospinning for adsorption of Cr<sup>3+</sup> from aqueous solution  
Ahmed A. Taha, Junlian Qiao, Fengting Li, Bingru Zhang ..... 610
- Removal of phosphate ions from aqueous solution using Tunisian clays minerals and synthetic zeolite  
Noureddine Hamdi, Ezzeddine Srasra ..... 617

**Atmospheric environment**

- Impacts of continuously regenerating trap and particle oxidation catalyst on the NO<sub>2</sub> and particulate matter emissions emitted from diesel engine  
Zhihua Liu, Yunshan Ge, Jianwei Tan, Chao He, Asad Naeem Shah, Yan Ding, Linxiao Yu, Wei Zhao ..... 624
- Dry deposition velocity of total suspended particles and meteorological influence in four locations in Guangzhou, China  
Leifu Chen, Shaolin Peng, Jingang Liu, Qianqian Hou ..... 632
- Synthesis, characterization and experimental investigation of Cu-BTC as CO<sub>2</sub> adsorbent from flue gas  
Jiangkun Xie, Naiqiang Yan, Zan Qu, Shijian Yang ..... 640
- Aerosol effects on ozone concentrations in Beijing: A model sensitivity study  
Jun Xu, Yuanhang Zhang, Shaoqing Zheng, Youjiang He ..... 645
- Measurement of air exchange rates in different indoor environments using continuous CO<sub>2</sub> sensors  
Yan You, Can Niu, Jian Zhou, Yating Liu, Zhipeng Bai, Jiefeng Zhang, Fei He, Nan Zhang ..... 657
- Influence of different weather events on concentrations of particulate matter with different sizes in Lanzhou, China  
Xinyuan Feng, Shigong Wang ..... 665

**Terrestrial environment**

- Sorption of chlorophenols onto fruit cuticles and potato periderm  
Yungui Li, Yingqing Deng, Baoliang Chen ..... 675
- Effects of urea and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> on nitrification and acidification of Ultisols from Southern China  
Deli Tong, Renkou Xu ..... 682
- Health risk assessment of heavy metals in soils and vegetables from wastewater irrigated area, Beijing-Tianjin city cluster, China  
Yanchun Wang, Min Qiao, Yunxia Liu, Yongguan Zhu ..... 690
- PCDD/Fs in soil around a hospital waste incinerator: comparison after three years of operation  
Xiaodong Li, Mi Yan, Jie Yang, Tong Chen, Shengyong Lu, Jianhua Yan ..... 699
- Dissolved organic sulfur in streams draining forested catchments in southern China  
Zhanyi Wang, Xiaoshan Zhang, Zhangwei Wang, Yi Zhang, Bingwen Li, Rolf Vogt ..... 704

**Environmental biology**

- Ammonium-dependent regulation of aerobic methane-consuming bacteria in landfill cover soil by leachate irrigation  
Fan Lü, Pinjing He, Min Guo, Na Yang, Liming Shao ..... 711
- Steady performance of a zero valent iron packed anaerobic reactor for azo dye wastewater treatment under variable influent quality  
Yaobin Zhang, Yiwen Liu, Yanwen Jing, Zhiqiang Zhao, Xie Qian ..... 720
- Identification of naphthalene metabolism by white rot fungus *Armillaria* sp. F022  
Tony Hadibarata, Abdull Rahim Mohd Yusoff, Azmi Aris, Risky Ayu Kristanti ..... 728

**Environmental health and toxicology**

- Inhibition of ROS elevation and damage to mitochondrial function prevents lead-induced neurotoxic effects on structures and functions of AFD neurons in *Caenorhabditis elegans*  
Qiuli Wu, Peidang Liu, Yinxia Li, Min Du, Xiaojuan Xing, Dayong Wang ..... 733

**Environmental catalysis and materials**

- Photodegradation of Norfloxacin in aqueous solution containing algae  
Junwei Zhang, Dafang Fu, Jilong Wu ..... 743
- Synthesis of TiO<sub>2</sub> nanoparticles in different thermal conditions and modeling its photocatalytic activity with artificial neural network  
Fatemeh Ghanbary, Nasser Modirshahla, Morteza Khosravi, Mohammad Ali Behnajady ..... 750
- Preparation of Fe<sub>x</sub>Ce<sub>1-x</sub>O<sub>y</sub> solid solution and its application in Pd-only three-way catalysts  
Jianqiang Wang, Meiqing Shen, Jun Wang, Mingshan Cui, Jidong Gao, Jie Ma, Shuangxi Liu ..... 757
- Dechlorination of chlorophenols by zero valent iron impregnated silica  
Praveena Juliya Dorathi, Palanivelu Kandasamy ..... 765
- Photocatalytic degradation of perfluorooctanoic acid with β-Ga<sub>2</sub>O<sub>3</sub> in anoxic aqueous solution  
Baoxiu Zhao, Mou Lv, Li Zhou ..... 774

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## Removal of arsenate and arsenite from aqueous solution by waste cast iron

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### Abstract

The removal of As(III) and As(V) from aqueous solution was investigated using waste cast iron, which is a byproduct of the iron casting process in foundries. Two types of waste cast iron were used in the experiment: grind precipitate dust (GPD) and cast iron shot (CIS). The X-ray diffraction analysis indicated the presence of Fe<sup>0</sup> on GPD and CIS. Batch experiments were performed under different concentrations of As(III) and As(V) and at various initial pH levels. Results showed that waste cast iron was effective in the removal of arsenic. The adsorption isotherm study indicated that the Langmuir isotherm was better than the Freundlich isotherm at describing the experimental result. In the adsorption of both As(III) and As(V), the adsorption capacity of GPD was greater than CIS, mainly due to the fact that GPD had higher surface area and weight percent of Fe than CIS. Results also indicated the removal of As(III) and As(V) by GPD and CIS was influenced by the initial solution pH, generally decreasing with increasing pH from 3.0 to 10.5. In addition, both GPD and CIS were more effective at the removal of As(III) than As(V) under given experimental conditions. This study demonstrates that waste cast iron has potential as a reactive material to treat wastewater and groundwater containing arsenic.

**Key words:** waste cast iron; arsenic removal; sorption; batch experiment

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### Introduction

Arsenic is a toxic trace element, existing in nature as arsenite (As(III)) and arsenate (As(V)) depending on the redox conditions. Under oxidizing environments, As(V) is predominant while As(III) occurs under reducing conditions. As(III) is known to be more toxic and mobile than As(V) (Giménez et al., 2007; Bang and Meng, 2004). In many countries such as Bangladesh, Chile, and India, arsenic occurs naturally in groundwater at concentrations exceeding 10 µg/L (guideline of World Health Organization), causing serious health-related problems and human mortality (Dixit and Hering, 2003).

Zero-valent iron (Fe<sup>0</sup>) is widely applied in arsenic removal filters and reactive barriers because it is nontoxic and inexpensive (Ahn et al., 2003; Leupin and Hug, 2005; Kanel et al., 2006). The interaction between arsenic and Fe<sup>0</sup> has been investigated by several researchers (Krishna et al., 2001; Ramaswami et al., 2001; Su and Puls, 2003, 2008; Kanel et al., 2005; Yu et al., 2006; Tyrovola et al., 2007; Cornejo et al., 2008; Katsoyiannis et al., 2008). These studies have investigated various aspects of As removal including: the effect of oxyanion competition on the removal kinetics of As(III) and As(V) (Su and Puls,

2001a), influence of iron corrosion rate on the removal rate of As(V) (Melitas et al., 2002a), redox reactions occurring on the surface of Fe<sup>0</sup> in arsenic solution using the electrochemical method (Melitas et al., 2002b), speciation of As(III) and As(V) after reaction with Fe<sup>0</sup> materials, iron corrosion products, and iron oxides (Manning et al., 2002), arsenic removal mechanisms and design factors for contaminated groundwater (Nikolaidis et al., 2003), arsenic removal during filtration through a mixture of iron filings and quartz sand (Leupin et al., 2005), removal of high-level As(III) in simulated groundwater (Lien and Wilkin, 2005), removal of As(V) and As(III) from geothermal waters in agricultural regions in the presence of nitrate and phosphate (Tyrovola et al., 2006), and removal of As(V) in the presence of various anions, manganese and organic matter (Biterna et al., 2007). These studies have shown that Fe<sup>0</sup> is effective at the removal of As(III) and As(V) from aqueous solutions, but the removal efficiency is influenced by various factors such as pH, temperature, redox conditions, and competitive anions. Further studies are, therefore, necessary to improve our knowledge regarding the interaction between arsenic and Fe<sup>0</sup>.

The objective of this study was to investigate the removal of As(III) and As(V) from aqueous solution by waste cast iron, which is a byproduct of the iron casting

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process in foundries. The characteristics of waste cast iron were elucidated using an X-ray fluorescence (XRF) spectrometer, an electron probe microscopic analyzer (EPMA), and a high resolution X-ray diffraction (HRXRD) spectrometer. Further, batch experiments were performed to determine removal effectiveness of waste cast iron for different concentrations of As(III) and As(V) and for various initial pH levels.

## 1 Materials and methods

### 1.1 Materials

Waste cast iron used in this study was obtained from the Macheon industrial complex in Jinhae, Korea. Two types of waste cast iron were used in the experiment: grind precipitate dust (GPD) and cast iron shot (CIS). GPD is a major form of waste generated from grinding and polishing cast iron products, while CIS is a byproduct of cast steel shot which is a ballast material used in the iron casting process. The stock solutions of As(V) and As(III) were prepared by dissolving reagent-grade sodium arsenate ( $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ , 98%+, Sigma-Aldrich, USA) and sodium arsenite ( $\text{NaAsO}_2$ , 0.05 mol/L (our response: confirmed), Merck, USA) in deionized water.

### 1.2 Characterization of waste cast iron

The chemical composition of waste cast iron was investigated using an XRF spectrometer (XRF-1700, Shimadzu Co., Japan). Surface area was determined by Brunauer-Emmett-Teller (BET)  $\text{N}_2$  adsorption analysis using a surface area analyzer (Coulter SA 3100, Coulter Co., USA). Images of waste cast iron were taken by using an EPMA (JXA-8900R, JEOL, Japan). The mineralogical and crystalline structural properties were examined using a cobalt target ( $\text{Co } K\alpha_1$ ) HRXRD spectrometer (MXP3A-HF22, MAC Science, Japan) with  $2\theta = 10\text{--}90^\circ$ . The point of zero charge ( $\text{pH}_{\text{pzc}}$ ) was determined by the potentiometric acid-base titration (Mörnstam et al., 1997).

### 1.3 Batch experiments

An adsorption isotherm study was conducted using batch test at pH 4.5. Five grams of GPD or CIS was added to 45 mL arsenic solution with various concentrations of As(V) or As(III) in polypropylene conical tubes. Ionic strength was adjusted by using 0.01 mol/L  $\text{NaNO}_3$ . Anaerobic conditions were maintained using a nitrogen purging device. The tubes were mixed at  $22^\circ\text{C}$  and 30 r/min using a hag rotator (FinePCR Co., USA). After 12 hr, the sample was taken, centrifuged, and filtered through a  $0.45\text{-}\mu\text{m}$  membrane filter. Then, the arsenic concentration

was measured by inductively coupled plasma-atomic emission spectroscopy (ICP-AES) (ICP-1000V, Shimadzu Co., Japan). To examine the effect of solution pH on removal, another batch study was performed in duplicate using the same procedures used in the isotherm study. The initial arsenic concentration was 3.3 mmol/L with the solution pH ranging from 3.0 to 10.5.

## 2 Results and discussion

### 2.1 Characteristics of waste cast iron

The characteristics of waste cast iron are summarized in Table 1. The particles sizes of the GPD and CIS used in the experiment were  $< 0.1$  and  $0.85\text{--}1.0$  mm, respectively. GPD had a surface area (BET) of  $2.1116\text{ m}^2/\text{g}$ , which was about four times larger than that of CIS ( $0.5824\text{ m}^2/\text{g}$ ). The chemical composition from XRF analysis showed that the major constituents of waste cast iron were Fe and Si. GPD and CIS contained 93.20% and 72.01% of Fe, respectively, indicating that the iron content of GPD was larger than that of CIS. EPMA images and patterns of waste cast iron are presented in Fig. 1. The EPMA patterns of GPD (Fig. 1b) and CIS (Fig. 1d) also demonstrated that Fe and Si were the major constituents of waste cast iron.

The images of GPD and CIS before and after reaction with arsenic solution are illustrated in Fig. 2. GPD and CIS changed to a yellow-brown color after reaction with As(III) and As(V). This phenomenon can be attributed to the anaerobic corrosion of  $\text{Fe}^0$  to iron corrosion products (iron oxides). In the corrosion process, water plays a significant role as an oxidant ( $2\text{H}_2\text{O} + \text{Fe}^0 = \text{Fe}^{2+} + \text{H}_2 + 2\text{OH}^-$ ) (Su and Puls, 2003; Farrell et al., 2001; Bang et al., 2005a). The XRD patterns of GPD and CIS before and after reaction with arsenic solution are presented in Fig. 3. Major peaks on the XRD of GPD indicated the presence of  $\text{Fe}^0$  (degree = 2.02, 1.43, 1.17), while major peaks on the XRD of CIS showed the presence of  $\text{Fe}^0$  along with quartz (degree = 3.34, 1.37). The XRD patterns after reaction with arsenic solution were similar to the patterns before reaction, indicating that no crystalline products form, rather amorphous and/or poorly crystalline hydrous ferric oxides ( $\text{FeOOH}$ ,  $\text{Fe}(\text{OH})_3$ ) were formed (Furukawa et al., 2002; Zhang et al., 2007).

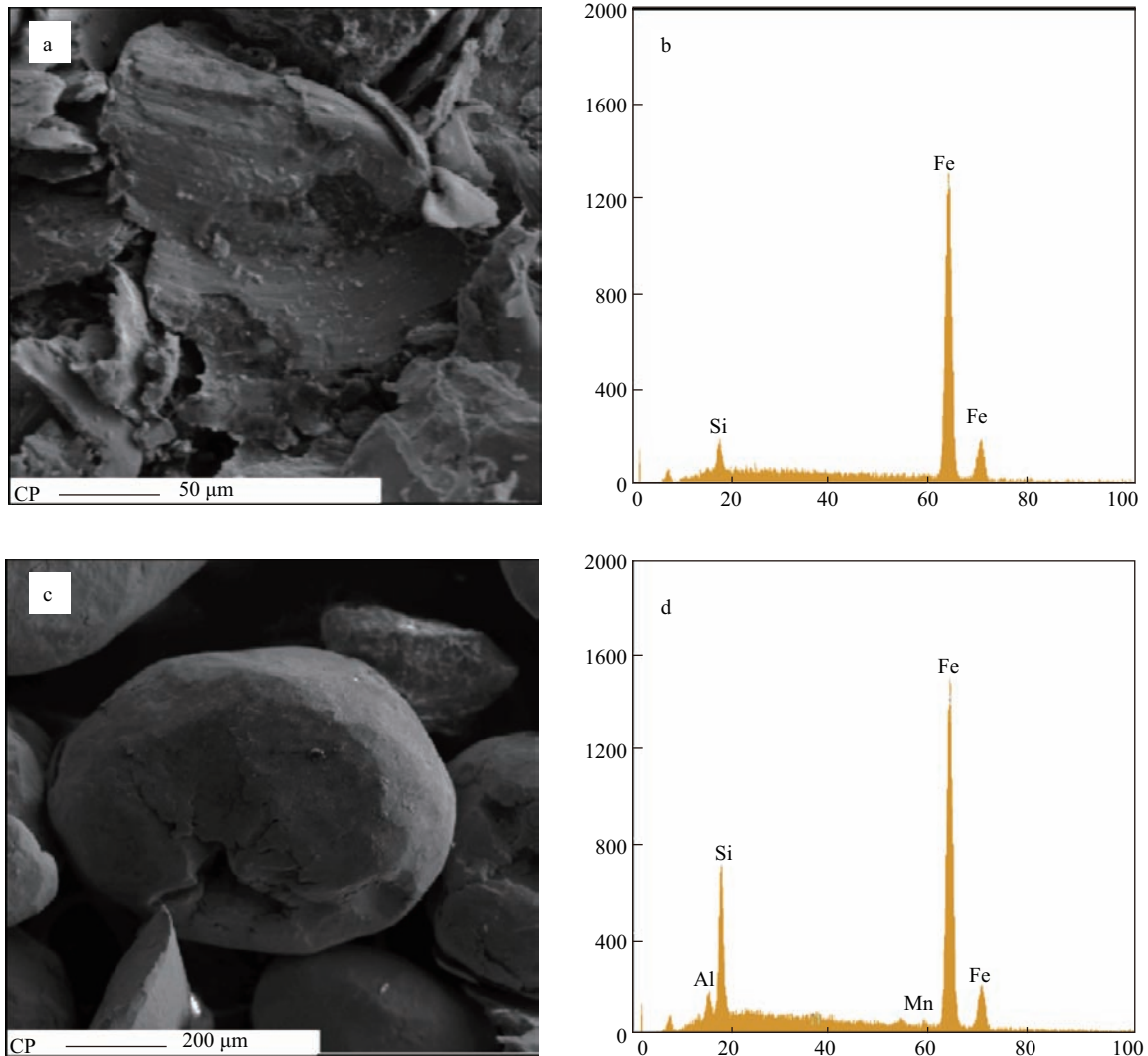
### 2.2 Equilibrium adsorption isotherms

Adsorption isotherms of As(III) and As(V) in GPD and CIS at an initial solution pH of 4.5 and ionic strength of 0.01 mol/L  $\text{NaNO}_3$  are presented in Fig. 4. The Langmuir and Freundlich isotherm models were used to analyze the

**Table 1** Characteristics of waste cast iron

	Particle size (mm)	Surface area ( $\text{m}^2/\text{g}$ )	$\text{SiO}_2$ (wt.%)	$\text{Al}_2\text{O}_3$ (wt.%)	$\text{TiO}_2$ (wt.%)	$\text{Fe}_2\text{O}_3$ (wt.%)	MnO (wt.%)	Total (wt.%)
GPD	$< 0.1$	2.1116	5.12	1.12	0.00	93.20	0.15	99.59
CIS	$0.85\text{--}1.0$	0.5824	25.84	0.95	0.00	72.01	0.60	99.40

GPD: grind precipitate dust; CIS: cast iron shot.



**Fig. 1** Waste cast iron used in the experiment. (a) electron probe microscopic analyzer (EPMA) image of GPD, (b) EPMA pattern of GPD, (c) EPMA image of CIS, (d) EPMA pattern of CIS.

adsorption data:

$$S = \frac{Q_{\max} K_L C}{1 + K_L C} \quad (1)$$

$$S = K_F C^{1/n} \quad (2)$$

where,  $S$  is the mass of solute adsorbed per unit mass of adsorbent,  $C$  is the concentration of solute in the aqueous solution at equilibrium,  $K_L$  is the Langmuir adsorption constant related to the binding energy,  $Q_{\max}$  is the maximum mass of solute adsorbed per unit mass of adsorbent (adsorption capacity),  $K_F$  is the distribution coefficient, and  $1/n$  is the Freundlich constant. Values of  $K_L$ ,  $Q_{\max}$ ,  $K_F$ , and  $1/n$  can be determined from the linear forms of Langmuir and Freundlich models.

The equilibrium isotherm constants of Freundlich and Langmuir models are given in Table 2. In the Freundlich model, the distribution coefficient ( $K_F$ ) was in the range from 0.937 to 1.149 L/g while the Freundlich constant ( $1/n$ ) was between 0.242 and 0.875. In the Langmuir model, the adsorption constant ( $K_L$ ) was in the range of 0.0034 to 0.0548 L/mg while the adsorption capacity ( $Q_{\max}$ ) was between 4.077 and 212.894 mg/g. As presented in Table 2, the correlation coefficients ( $R^2$ ) of the Langmuir model were greater than those of Freundlich model, indicating that the Langmuir isotherm was better than Freundlich isotherm at describing the experimental results. In the Langmuir model, monolayer adsorption is assumed to occur at a homogeneous surface with a finite number of adsorption sites. In monolayer adsorption, all

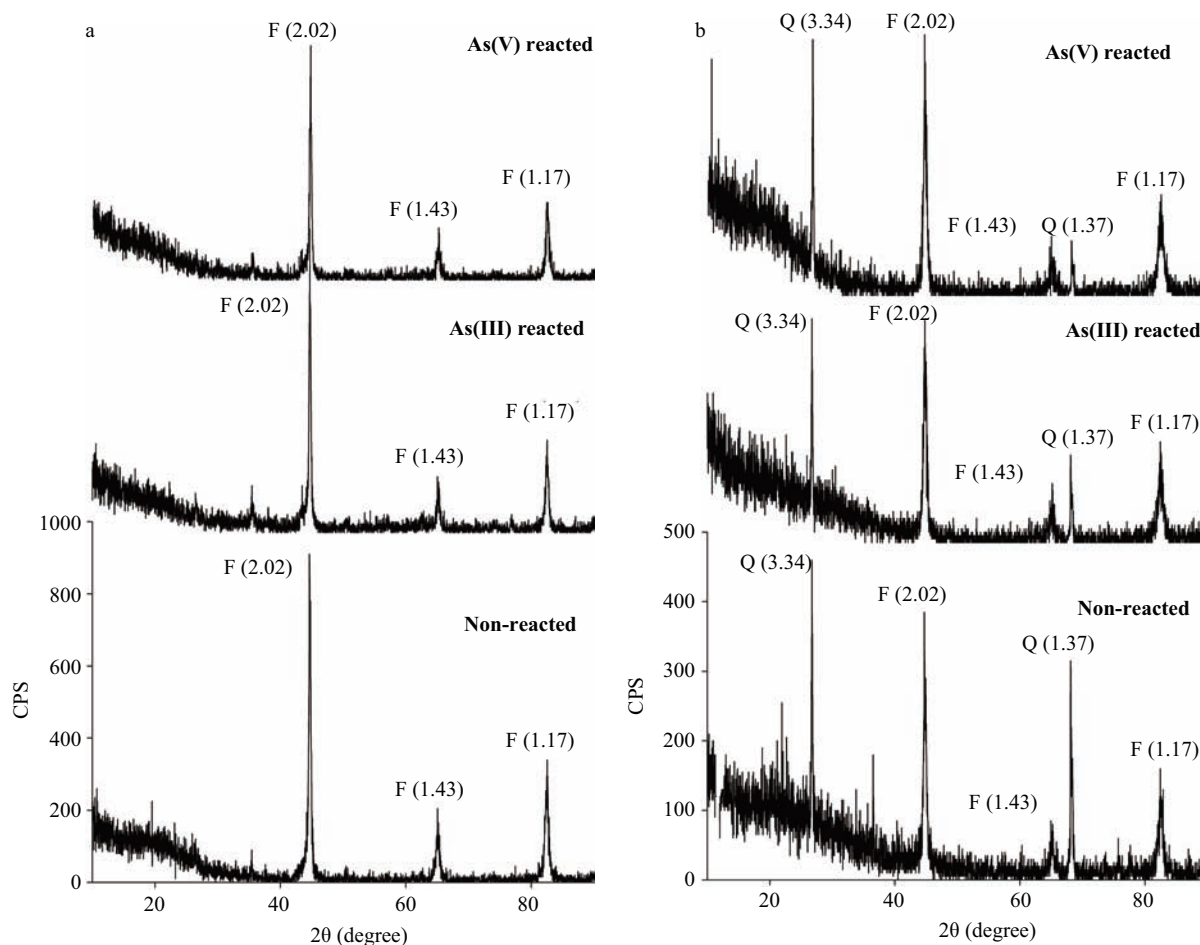
**Table 2** Equilibrium isotherm constants for waste cast iron

	Freundlich isotherm model			Langmuir isotherm model		
	$K_F$ (L/g)	$1/n$	$R^2$	$K_L$ (L/mg)	$Q_{\max}$ (mg/g)	$R^2$
As(V) GPD	0.937	0.653	0.952	0.0034	82.441	0.960
As(III) GPD	1.149	0.875	0.880	0.0085	212.894	0.993
As(V) CIS	0.955	0.242	0.871	0.0548	4.077	0.935
As(III) CIS	1.023	0.580	0.962	0.0174	23.833	0.978





**Fig. 2** GPD and CIS before and after reaction with arsenic solution. (a) non-reacted GPD; (b) As(V)-reacted GPD; (c) As(III)-reacted GPD; (d) non-reacted CIS; (e) As(V)-reacted CIS; (f) As(III)-reacted CIS.

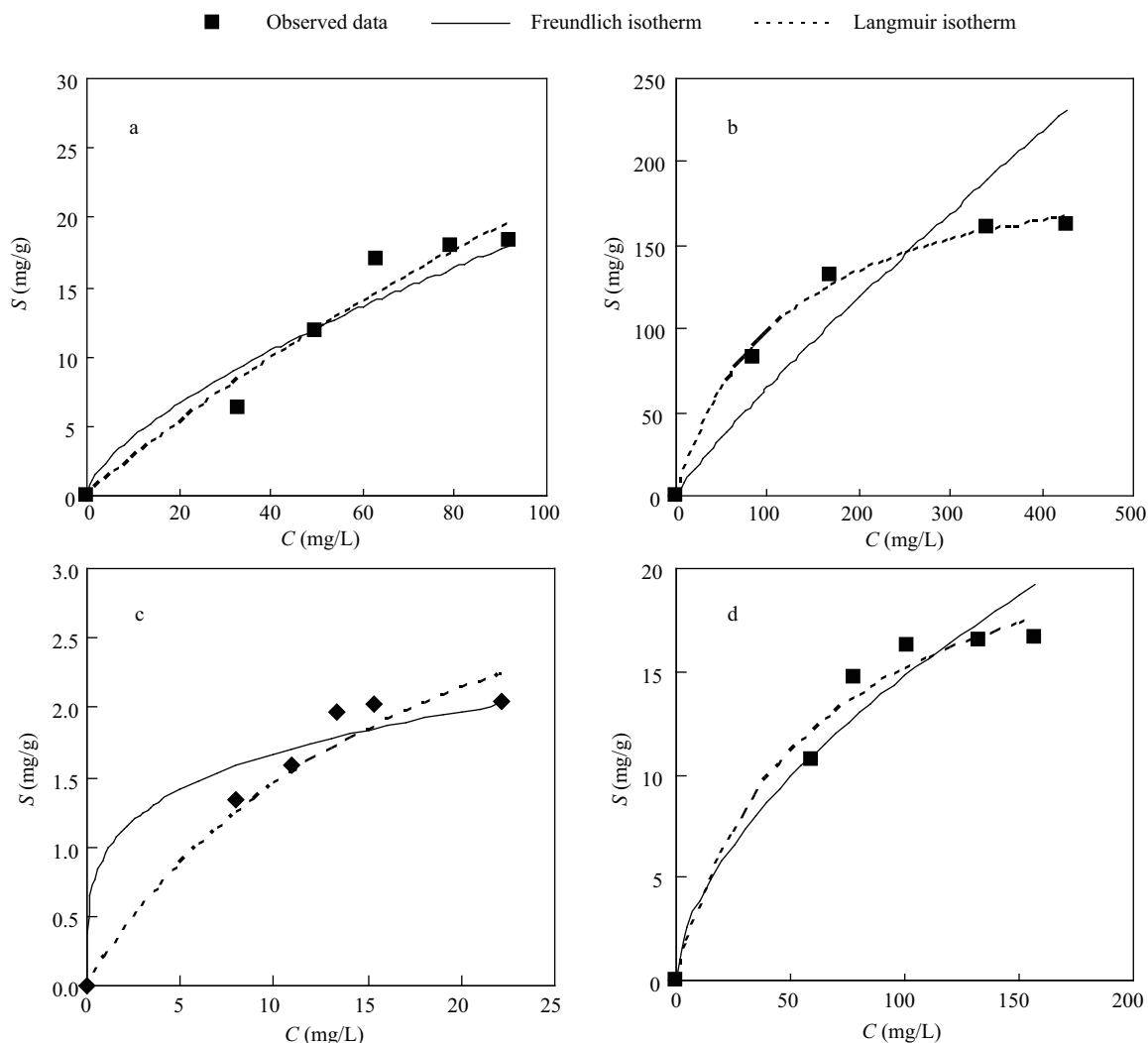


**Fig. 3** X-ray diffraction (XRD) patterns of GPD (a) and CIS (b). Q = quartz, F = Fe<sup>0</sup>.

the adsorbed molecules are in contact with the surface layer of the adsorbent (Jeon et al., 2009).

In the adsorption of As(V), the solution pH increased to 10.0 and 10.1 (equilibrium pH) in GPD and CIS, respectively. The value of  $Q_{\max}$  in GPD was 82.441 mg/g, which was about 20 times larger than the value in CIS

(4.077 mg/g). Also, the solution pH increased to 9.0 and 8.8 (equilibrium pH) in GPD and CIS, respectively, in the adsorption of As(III). The value of  $Q_{\max}$  in GPD (212.894 mg/g) was about 10 times larger than the value in CIS (23.833 mg/g). This result can be attributed to different physical and chemical characteristics between GPD and

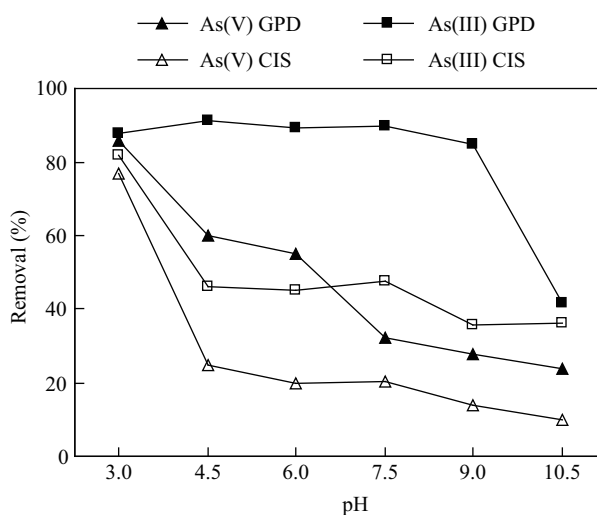


**Fig. 4** Adsorption isotherms of As(III) and As(V) in GPD and CIS. pH = 4.5; ionic strength = 0.01 mol/L NaNO<sub>3</sub>. (a) As(V) in GPD; (b) As(III) in GPD; (c) As(V) in CIS; (d) As(III) in CIS.

CIS. Among the factors, surface area and Fe content might be the major ones. GPD had higher surface area (2.1116 m<sup>2</sup>/g) than CIS (0.5824 m<sup>2</sup>/g) and also that the chemical composition, based on XRF analysis, indicated that GPD had higher weight percent of Fe (93.20%) than CIS (72.01%).

### 2.3 Effect of pH

The effect of initial solution pH on the average removal of As(III) and As(V) by GPD and CIS at an ionic strength of 0.01 mol/L NaNO<sub>3</sub> is presented in Fig. 5. Arsenic removal by waste cast iron was clearly affected by the solution pH. In the adsorption of As(V) to GPD, the removal percent gradually decreased from 86% to 24% with increasing pH from 3.0 to 10.5. In the case of As(V) adsorption to CIS, the removal percent decreased rapidly from 77% to 25% between pH 3.0 and 4.5 and decreased gradually thereafter to 10% at pH 10.5. In the adsorption of As(III) to GPD, the removal percent remained relatively constant, at around 90% for pH 3.0–9.0 and then dropped sharply to 42% at pH 10.5. In the case of As(III) adsorption to CIS, the removal percent dropped rapidly from 82% to 46% between pH 3.0 and 4.5 and decreased gradually thereafter to 36% at pH

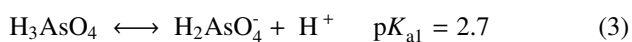


**Fig. 5** Average removal of As(III) and As(V) by GPD and CIS for solution pH ranging from 3.0 to 10.5. Ionic strength = 0.01 mol/L NaNO<sub>3</sub>.

10.5. For the same pH conditions, the removal of As(III) and As(V) by GPD was greater than by CIS. This result can be attributed to surface area and chemical composition mentioned previously.

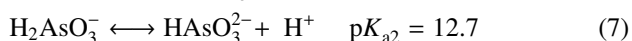
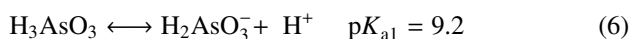
The pH dependency of adsorption of As(III) and As(V)

to waste cast iron is primarily related to ionization of arsenic and adsorbent in accordance with pH change. The speciation of arsenic acid ( $\text{H}_3\text{AsO}_4$ , As(V)) is influenced by the solution pH (ionic strength = 0.01 mol/L):



In the pH range between 2.7 and 6.8,  $\text{H}_2\text{AsO}_4^-$  (monovalent anion) is dominant, and the iron oxide surfaces have a net positive charge ( $\text{pH}_{\text{pzc}}$ : GPD = 7.57; CIS = 7.16). In this pH range, the adsorption of As(V) is favorable even though the percent removal gradually decreased with increasing pH. For pH values of 6.8–11.6,  $\text{HAsO}_4^{2-}$  (divalent anion) dominates, and the surface charges of iron oxides change from positive to negative (Katsoyiannis et al., 2008). Thus, the adsorption of As(V) is unfavorable in this pH range since electrostatic interaction between As(V) and oxide surface becomes repulsive (Su and Puls, 2001b). Therefore, the percent removal further decreases with increasing pH.

The speciation of arsenious acid ( $\text{H}_3\text{AsO}_3$ , As(III)) is also affected by the solution pH (ionic strength = 0.01 mol/L):



$\text{H}_3\text{AsO}_3$  (uncharged form) dominates at pH 2.0–9.2, and the adsorption of this species to GPD may occur as inner-sphere bidentate surface complexes with iron oxide surfaces (Manning et al., 2002; Stachowicz et al., 2006).  $\text{H}_2\text{AsO}_3^-$  (monovalent anion) is dominant at pH 9.2–12.7. The adsorption of As(III) in this pH range is unfavorable. The negatively-charged monovalent anion becomes predominant, and the oxide surface becomes negatively charged, resulting in electrostatic repulsion between As(III) and oxide surface (Giménez et al., 2007). As presented in Fig. 5, the removal curve for As(III) by CIS is somewhat different from the case of As(III) by GPD. In the case of As(III) adsorption to CIS, the removal percent dropped rapidly under acidic conditions (pH 3.0–4.5) and decreased gradually thereafter. This discrepancy may be attributed to differences in particle size and  $\text{Fe}_2\text{O}_3$  content between CIS and GPD.

#### 2.4 Removal of As(III) and As(V)

In GPD, the adsorption capacity ( $Q_{\text{max}}$ ) of As(III) was 212.894 mg/g, which was about 2.5 times greater than that of As(V) (82.441 mg/g). Also, the  $Q_{\text{max}}$  value of As(III) in CIS (23.833 mg/g) was about six times greater than that of As(V) (4.077 mg/g). In addition, percent removal of As(III) in both GPD and CIS were greater than that of As(V) in the pH range between 3.0 and 10.5. Percent removal of As(III) at pH 6.0 in GPD was 89.5% while percent removal of As(V) was 55.0%. In CIS, percent removal of As(III) at pH 6.0 was 45.0%, which is greater than that of As(V) (20.0%).

Our results indicated that waste cast iron was more effective at the removal of As(III) than As(V) under given experimental conditions. Other researchers have also reported that  $\text{Fe}^0$  had different removal efficiencies between As(III) and As(V). Su and Puls (2001b) showed with batch experiments that As(III) had greater removal rates than As(V) in Fisher and Peerless  $\text{Fe}^0$ . Bang et al. (2005b) reported that under nitrogen at pH 4 and 7, As(III) removal was greater than As(V) removal after 5 days of batch reactions. Sun et al. (2006) showed that As(V) was removed more efficiently than As(III) at pH 8.28 in an aerobic batch system. They reported, however, that As(III) removal was faster than As(V) removal in a relatively anaerobic system. Tyrovola et al. (2006) reported that As(V) had greater removal rate than As(III) in experimental temperatures between 20 and 40°C. These studies demonstrate that researchers have reported different results regarding the difference in the removal efficiency between As(III) and As(V). These discrepancies may be attributed to the different experimental conditions including variations in type of  $\text{Fe}^0$ , solution pH/composition, and redox conditions.

### 3 Conclusions

Two types of waste cast iron, GPD and CIS, were used to remove As(III) and As(V) in batch experiments. Results showed that waste cast iron was effective in the removal of arsenic from aqueous solution. The adsorption isotherm study indicated that the Langmuir isotherm was better than the Freundlich isotherm at describing the experimental result. In the adsorption of both As(III) and As(V), the adsorption capacity ( $Q_{\text{max}}$ ) of GPD was greater than CIS, mainly due to the fact that GPD had higher surface area and weight percent of Fe than CIS. Results also indicated the removal of As(III) and As(V) by GPD and CIS was influenced by the initial solution pH, generally decreasing with increasing pH from 3.0 to 10.5. In addition, both GPD and CIS were more effective at the removal of As(III) than As(V) under given experimental conditions. This study demonstrates that waste cast iron, an industrial byproduct, has potential as a reactive material to treat wastewater and groundwater containing arsenic. More experiments should be performed under natural water conditions to examine the effects of natural factors (e.g. heavy metals such as  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Cr}^{6+}$ ) on the arsenic adsorption to waste cast iron and further test its performance in relation to arsenic removal.

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