

Adsorption Characteristics of Aqueous Arsenic(III) and Arsenic(V) in Taiwan Soils

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Abstract: This study investigate the adsorption characteristics of aqueous arsenate [As(V)] and arsenite [As(III)] in Taiwan soils namely pinchen, chunliao and baiho. The results from the batch adsorption experiments showed that the adsorptive capability of the soils increased with increasing amounts of iron (Fe), manganese (Mn) and aluminum (Al) oxides. In the tested soils, the adsorption of As(V) was higher than that of As(III). The concentration of phosphate in tested soils negatively correlated with the adsorption of aqueous arsenic species As(V) and As(III). The desorption of arsenic occurred within 2–3 h after soils were submerged in water as evidenced by the increased concentration of As(V) and As(III) in the reaction systems. The kinetics analyses of the adsorption of As(III) and As(V) by the soils showed that the major part of reaction was of zero order, while minor part followed 3rd order reaction. This fundamental study will be helpful for further application in the treatment of arsenic contaminated Taiwan soils.

Keywords: Aqueous as(V) and as(III); taiwan Ssoil; adsorption; kinetics.

1. Introduction

Arsenic is one of the most harmful and toxic element found in nature. It is a slow poisoning element which severely affects the human health and other living organisms [1]. Naturally arsenic is found in rocks and minerals. Due to weathering and erosion of rocks and soils and volcanic emissions, arsenic is in contact with the ground water and creates pollution [2]. Apart from natural sources, arsenic contamination is also associated with anthropogenic activities, from application of agricultural pesticides, mining, and disposal of industrial chemical waste, land filling of sewage sludge and

burning of fossil fuels [2]. Industrially arsenic is mainly used as a wood preservative and hence, it has been used in dyes, paints and pigmenting substances. It is also used in glass-making, electronics manufacturing and leather tanning industries. A small amount of arsenic is used in both human and animal medications and care products, and it is present in many food supplement products also [2]. The actual distribution, mobility, bioavailability, toxicity, bioaccumulation, and biodegradability of arsenic depend not only on its total concentration but also on its

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Received 29 June 2012
Revised 10 August 2012
Accepted 22 October 2012

chemical form in the sample [2]. Arsenic can exist in inorganic form, organic form and gaseous state. Organic arsenic species are less harmful to health and readily eliminated by the body. However, an inorganic form of arsenic is highly toxic compared to organic arsenic. Inorganic arsenate and arsenite referred to a pentavalent As(V) and trivalent As(III), respectively, are most common in natural waters [3]. As(III) is 60 times more toxic than As(V), due to its greater cellular uptake [3].

Drinking waters normally derive from surface water or groundwater, depending on local availability, which are very variable in their arsenic contents, although the highest concentrations are often found in groundwater [4]. The arsenic contamination of soils and water represents a great threat to human health due to its high potential to enter into the food chain [5]. Long-term exposure to inorganic arsenic compounds can lead to various diseases such as conjunctivitis, hyperkeratosis, hyperpigmentation, cardiovascular diseases, disorders of the central nervous system and peripheral vascular system, skin cancer and gangrene of the limbs [5–7]. Additionally, arsenic poisoning also leads to death from multisystem organ failure by allosteric inhibition of sulfhydryl containing essential metabolic enzymes. It also interferes with some biochemical processes involving phosphorous due to the similarity in their chemical properties [5–7].

The presence of arsenic in the environment is regulated by many environmental and public health agencies or authorities. According to World Health Organization (WHO) and United States Environmental Protection Agency (US EPA), the maximum allowed concentration of arsenic in drinking water is $10 \mu\text{g L}^{-1}$ [8]. However, arsenic concentrations, about 100 times more than the permissible limit, are found in many parts of the world [2, 4]. The

rapid industrialization in Taiwan results in difficulties in land acquisition. As a result, industrial parks have been built around agricultural areas, and caused pollution problems. To increase the agricultural output, farmers use pesticides and fertilizer excessively, which are another arsenic pollution source. Blackfoot disease that prevails in the southwest coast of Taiwan is an endemic disease cause by arsenic intoxication [9].

Adsorption is one of the most commonly reported, and possibly the initial reaction to occur when arsenic interacts with soils [10]. There are many factors which affects the arsenic adsorption in soil. Arsenic has been known to strongly adsorb to inorganic constituents in soils such as clay minerals and iron (Fe) and aluminum (Al) oxides/hydroxides. Both As(V) and As(III) have strong affinities for Fe and Al oxides [10, 11]. From adsorption study performed on 18 soils with varying soil properties, Burns et al. (2006) reported that arsenic adsorption was highly correlated with the soil pH and iron oxide content [12]. In addition, X-ray spectroscopy studies demonstrated the formation of strong inner-sphere complexes between adsorbed arsenic and the adsorbent surfaces that were mainly composed of Fe/Al oxides [10, 11]. Additionally, the concentration of phosphate (PO_4) is also a main factor which affects the As adsorption negatively, because PO_4 has a competition for adsorption sites with As(V) [13]. Understanding of the As(V) and As(III) adsorption processes in Taiwan soils is poor. Therefore, the objectives of this study were to examine the As(V) and As(III) adsorption behavior with soils samples collected around Taiwan. This fundamental study will be helpful for further application in the treatment of arsenic contaminated Taiwan soils.

2. Materials and methods

2.1. Soil sample collection

Alluvial soil samples with low arsenic concentration were collected from soil series of Chunliao in Sihu, Changhua and Baiho series from Guiren County, Taiwan. Red soil samples collected from soil series of Pinchen in Pusing, Taoyuan were used for the batch of adsorbability test for inorganic arsenic.

At every sampling site, ten points of soil sample were collected randomly, and every point of soil sample included surface soil (0–15 cm) and bottom soil (15–30 cm). The soil sample for the batch of adsorbability test was collected from surface soil (0–15 cm). Ten surface or bottom soil samples were mixed, air dried, grinded, and stored in plastic bottles after passed through 2 mm sieves.

2.2. Analysis of soil characteristics

The pH of the soil was measured in soil suspension with ratio of soil: water ratio of 1:1 (w/v) using glass electrode [14]. Electrical conductivity (EC) of the soil was measured with electric conductor using the soil suspension prepared as described above. Organic carbon was determined by dry heating method [15]. Soil texture was tested with the hydrometer method [16]. Cation exchange capacity of the soil was evaluated with the ammonium saturation method [17]. Valid phosphorous was determined with Bray NO.1 method [18]. Crystallized manganese, iron and aluminum oxides were determined using the methods of Mehra and Jackson (1960) [19]. CBD reagent solution was used to dissolve oxidized manganese, oxidized iron and oxidized aluminum in the soil and then the concentration of manganese, iron and aluminum in suspension was analyzed using Inductively Coupled Plasma-Optical Emission

Spectrometer (ICP–OES), in order to calculate the concentration of manganese, iron and aluminum in soil. Irregular iron, manganese and aluminum oxides extracted by ammonium oxalate were analyzed by Schwertmann (1964) method [20], using 0.2 M of ammonium oxalate at pH 3.0 for extraction.

2.3. Batch experiments on the adsorbability of arsenic based on soil compositions

Surface soil samples (with low arsenic content) of 10 g each were collected from three areas, of which two were alluvial and one was red soil, namely, sand shale alluvial from Guiren Tainan (baiho series), clay slate from Sihu Changhua (chunliao series), and red soil from Pusing Taoyuan (pinchen series). Each sample was added with 100 mL of HAsO_4^{2-} or AsO_2^- solution (As concentration is 6 mg L^{-1}) to mix into suspension with soil and solution ratio at 1:10 (w/v). Then the samples were oscillated for 1, 2, 4, 8, 12, 16 and 24 h, and centrifuged (33,000 g), the supernatant was filtered (No. 42) and measured the residue concentration of arsenic using ICP.

3. Results and discussion

The As(III) and As(V) adsorption tendency in Taiwan soils were shown in Figure 1–3. The adsorption follows in the order: pinchen > baiho > chunliao soil. It was interesting to note that pinchen soil series contain the higher content of oxides as well as lower content of phosphorous (P) as compared with chunliao and baiho soil series (Table 1). In our study, the P contents in soil samples collected from pinchen, baiho and chunliao were 6.78 mg kg^{-1} and 39.9 mg kg^{-1} and 228 mg kg^{-1} , respectively. Thus, pinchen and baiho soil provided less valid adsorptive positions for P than chunliao soil. These results indicate that the

tested soils can provide valid adsorptive positions to P and arsenic (Figure 1-3, Table 1). Violante and Pigna (2002) studied the competitive sorption of PO_4^{3-} and As(V) on selected clay minerals [13]. They found that PO_4^{3-} could inhibit As(V) sorption on clay minerals such as gibbsite and kaolinite. Smith et al. (2002) observed that the presence of PO_4^{3-} greatly decreased As(V) sorption by soils containing low amounts of

Fe oxides but had little effect on the amount of As(V) adsorbed by soils with high Fe content [21]. Goldberg (2002) found that there was no competitive effect of the presence of equimolar As(III) on As(V) adsorption [22]. Our results were in agreement with these previous reports that the P content negatively correlates with arsenic adsorption.

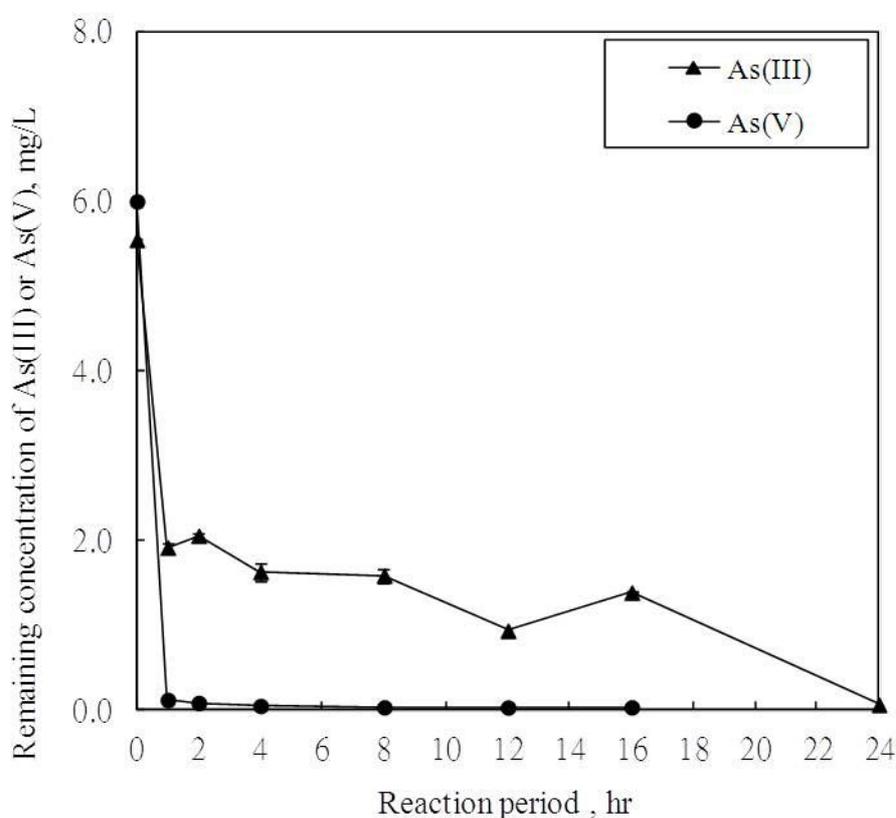


Figure 1. The adsorption tendency of aqueous As(III) and As(V) on pinchen soil series at room temperature

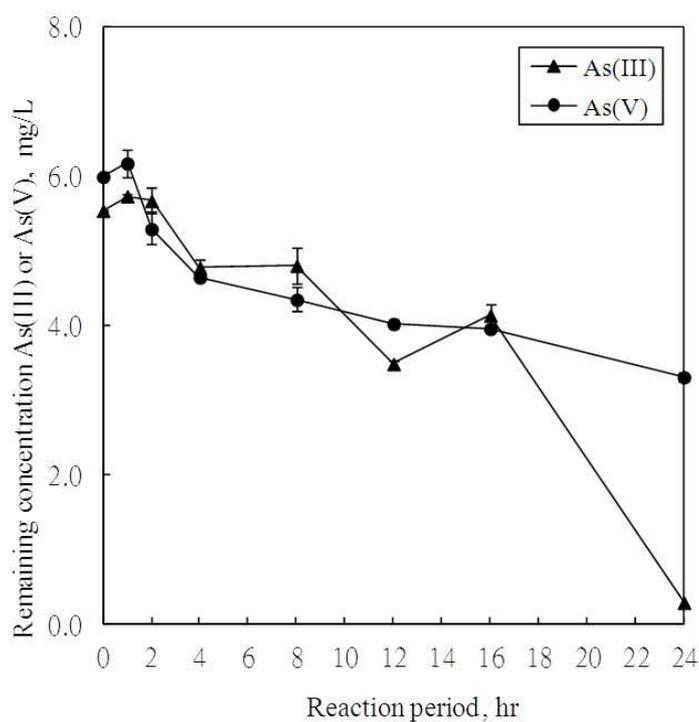


Figure 2. The adsorption tendency of aqueous As(III) and As(V) on chunliao soil series at room temperature

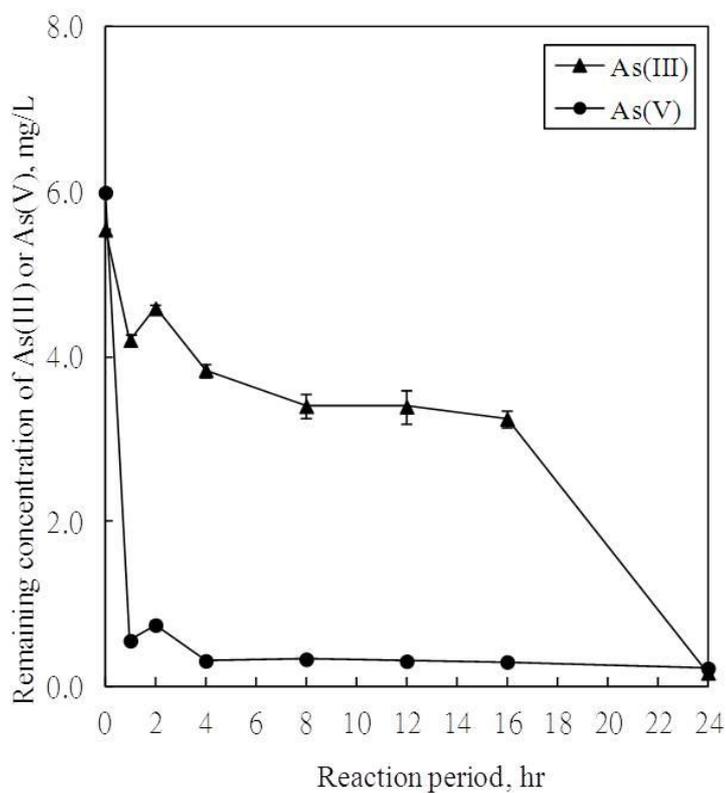


Figure 3. The adsorption tendency of aqueous As(III) and As(V) on baiho soil series at room temperature

As seen in Figure 1–3 and in Table 2, the concentration of As(III) and As(V) in water soluble form of surface soil collected from pinchen and baiho decreased rapidly in 0–1 h. In particular, the concentration of As(V) decreased from 6.01 mg kg⁻¹ to 0.12 mg kg⁻¹, and to 0.55 mg kg⁻¹, respectively. During the same period, the content of As(III) decreased in pinchen and baiho soils from 5.54 mg/kg to 1.91 mg kg⁻¹ and, to 4.20 mg kg⁻¹, respectively. These results indicate that as soon as arsenic enters into the soil, it was adsorbed by iron, aluminum and manganese oxides in soil, and this leads to a rapid drop in concentration of As(III) and As(V) in solution (Table 2). In addition, our data also indicate that the concentration of As(V) in pinchen and baiho soils remained nearly stable in from 1 h to 24 h, while the same for As(III) in 1 h to 16 h. This may due to the occupation of valid adsorption positions by As(III) and As(V) within 0–1 h, thus, results in slowdown of adsorption speed. Previously, it is reported that that the adsorption of As(V) from aqueous solution was rapid in the initial stages of contact and reached a maximum in the range of 35–60 min for soil [23]. In our study, within 0–1 h the residual concentration of As(V) in solution is lower than As(III) (Table 2). Therefore, the adsorption of As(V) is higher than that of As(III). Thus, the results of our study were in agreement with the results reported earlier [24, 25]. The oxidation state of As depends primarily on pH and redox conditions, with As(V) being the most stable form under aerobic conditions as the pH-dependent deprotonated oxyanions of arsenic acid (H₂AsO₄⁻ and HAsO₄²⁻) and As(III) the chemically dominant forms in reducing environment as a neutral species (i.e., pK_{a1} = 9.2) at natural pH. Therefore, the As(III) is more difficult to remove from water at neutral pH by means of adsorption and coprecipitation due to the lack of electrostatic attraction [26]. However, most As-enriched groundwater is generally

dominated by As(III), up to 96% of total As [27]. McGeehan and Naylor (1994) reported that the Fe and Mn oxides were restored and dissolved by the release of adsorbed arsenic to the solution [26]. Barrachina et al. (1996) observed that 80% of the total amount of As(III) adsorbed was sorbed by Spanish soil in the first 30 min [27]. O'Reilly et al. (2001) discovered that As(V) sorption on goethite was initially rapid, with over 93% As(V) adsorption within 24 h [28]. In this study, the concentration of As(III) in pinchen and baiho soils decreased rapidly from 16 h to 24 h. The reason was suspected that the soil saturated in water at an anaerobic state, so it restores iron and Mn, and the restoration of Mn increases the As(III) adsorption and it was oxidized into As(V). In addition, the As(III) in pinchen soil and, As(III) and As(V) in baiho soil increased the concentration after immersed in water for two hours, this may due to a result of arsenic desorption (Figure 1 and 3). Further studies were required to examine the kinetics of both As(III) and As(V) adsorption in the same soil system. This information is essential because both As(III) (predominantly in the reduced condition) and As(V) (predominantly in the oxidized condition) are often discovered in either redox environments because of the relatively slow redox transformation of As [29].

The adsorption of As(III) and As(V) by chunliao soil during the period of 0–16 h was much lower (Figure 2), as compared with pinchen and baiho soil (Figure 1 and 3). These results may possibly due to the relationship between the factors of valid adsorption position and oxide content. Another explanation was the relationship between pH and pE. The pH of chunliao soil was 7.21 and increased when the soil immersed in water, while the pE drops noticeably. During this time, As(V) may undergo reduction and transformed into As(III) in the reaction system.

Table 1. Physical and chemical properties of the soils for the adsorption of batch experiments

Sample site	Soil series	Texture	pH	EC	CEC	Org-C	Total As	Bray-I P	Citrate-bicarbonate-dithionite extractable			Oxalate extractable		
									Fe	Al	Mn	Fe	Al	Mn
				mS cm ⁻¹	cmol kg ⁻¹	%	— mg kg ⁻¹	—	— %	—	— mg kg ⁻¹	— %	—	— mg kg ⁻¹
Puhsin, Taoyuao	Pinchen	C	4.25	0.219	7.61	0.72	9.5	6.78	2.38	0.45	178	0.28	0.21	3.0
Hsihu, Changhua	Chunliao	LS	7.21	0.189	4.54	0.46	7.8	228	0.52	0.04	182	0.27	0.04	7.5
Guiren, Tainan	Baiho	L	4.82	0.626	8.61	0.74	8.0	39.9	0.68	0.09	112	0.60	0.08	4.5

Table 2. After the absorption of aqueous As(III) and As(V) by the surface soils, the remaining concentrations of As(III) and As(V) in the reaction systems at various reaction periods

Reaction period (h)	(Pinchen)		(Chunliao)		(Baiho)	
	As(III)	As(V)	As(III)	As(V)	As(III)	As(V)
	mg kg ⁻¹					
0	5.54±0.01 ^a	6.01±0.03	5.54±0.01	6.01±0.03	5.54±0.01	6.01±0.03
1	1.91±0.03	0.12±0.01	5.74±0.02	6.18±0.10	4.20±0.03	0.55±0.02
2	2.05±0.01	0.08±0	5.67±0.10	5.30±0.14	4.59±0.02	0.74±0.03
4	1.63±0.06	0.05±0.01	4.78±0.06	4.65±0.01	3.84±0.05	0.31±0.02
8	1.58±0.05	0.03±0	4.81±0.14	4.35±0.09	3.40±0.08	0.33±0
12	0.93±0	0.03±0	3.49±0.03	4.03±0.03	3.39±0.12	0.31±0
16	1.38±0.01	0.03±0	4.14±0.08	3.96±0.01	3.25±0.06	0.30±0
24	0.06±0.01	ND ^b	0.29±0.01	3.31±0.03	0.16±0.01	0.22±0.02

^aStandard deviation of the mean. ^bNot detectable

Table 3. The kinetic analyses of aqueous As(III) and As(V) absorbed by various soils

Soil series	Reaction period (h)	Reaction order	Correlation coefficient (R ²)	Rate constant (k)
As(III)				
Pinchen	0 - 1	0	1.000	3.630 ^a
	1 - 24	0	0.921	0.075 ^a
Chunliao	0 - 16	0	0.880	0.123 ^a
	16 - 24	0	1.000	0.481 ^a
Baiho	0 - 16	3	0.900	0.002 ^b
	16 - 24	0	1.000	0.386 ^a
As(V)				
Pinchen	0 - 1	0	1.000	5.880 ^a
	1 - 24	0	0.994	0.002 ^a
Chunliao	0 - 4	0	0.933	0.382 ^a
	4 - 24	0	0.990	0.063 ^a
Baiho	0 - 1	0	1.000	5.443 ^a
	1 - 24	3	0.895	0.414 ^b

a,b The units for the rate constants indicated are mg L⁻¹ h⁻¹ and [(mg L⁻¹)²]-1h⁻¹, respective

On the other hand, a few researchers studied the adsorption of As on the soil minerals (oxides, hydroxides, or clays) and found that adsorption decreased with increasing pH [25]. Desorption of As(III) and As(V) occurred after oscillation for 1 h (Figure 2). In 16–24 h of reaction time, the concentration of As(III) in chunliao soil dropped to near zero (Figure 2), may be due to chunliao alluvial soil contains higher amount of carbonate. Sadiq (1997) pointed out that arsenic in alkaline soil is adsorbed by carbonate, and arsenic and calcium arsenate formed by calcium ions, result in decrease in As(III) [30]. Previous reports about pH influence on As adsorption suggested that As(III) sorption was maximum at around pH 7 [31, 32], whereas As(V) sorption reached to maximum sorption around pH 4–7 and then decreased with increasing pH [22, 33]. However, further studies required to know the reason for the concentration of As(V) was not decreased within 16–24 h.

The evaluation on the As(III) and As(V) adsorbabilities of surface soil collected from pinchen, chunliao, and baiho were calculated with the following dynamics equation according to different reaction level simulated at different time blocks.

$$-\frac{dC}{dt} = kC^n \quad (1)$$

Where C, t, k, and n were represents the concentration, reaction time, rate constant and reaction order, respectively. The equations of zero and third degree are as follows:

$$C = C_0 - kt \quad (2)$$

$$\frac{1}{C^2} = \frac{1}{C_0^2} + 2kt \quad (3)$$

Where C_0 is the initial concentration.

Generally, within two adjacent test times, sample with rapid change in concentration was treated as 0 order reactions to calculate its rate constant. In consecutive times, the trend of concentration change was simulated

from order 0 to 3, and the highest reaction order was selected to simulate the optimal order. The results were shown in Table 3.

In 0–24 h, pinchen soil was of 0 order reaction to As(III) and As(V). In 0–1 h, the rate constant of As(V) adsorption was higher than that of As(III). In 1–24 h, As(III) was higher than As(V) (Figure 1 and Table 3), which are due to oxidization of As(III) into As(V). In chunliao soil system, the rate constant of As(V) in 0–4 h was five times larger than that in 4–24 h; the reason may be due to during this time As(V) not yet transformed into As(III). In 4–24 h, As(V) gradually transforms into As(III) so the reaction speed drops. As(III), in 16–24 h, increased 4 times because As(III) and calcium arsenate produced by calcium ion is settled (Figure 2 and Table 3). As(III) adsorption into baiho soil showed 3rd order reaction in 0–16 h, and 0 order reaction in 16–24 h. As(V) adsorption showed level 0 reaction in 0–1 h, and 3rd order reaction in 1–24 h (Figure 3 and Table 3). The reason for direct proportional reaction between reaction speed and concentration in 0–16 h and 1–24 h was yet to be studied. As seen in Table 3, most soil samples showed 0 order reaction to As(III) or As(V). Thus, the adsorption reaction rate has no correlation with arsenic concentration. Any reaction over order 0 should be caused by different compositions of soil.

4. Conclusions

In conclusion, this study demonstrate that the tested Taiwan soil pinchen, chunliao and baiho series with high content of oxides have high As(III) and As(V) adsorption capacity. Chunliao soil with higher content phosphate can compete for adsorptive position with arsenic resulted the reduced adsorption of As(III) and As(V). The adsorption of As(V) was higher as compared with As(III) by the tested three soil series. Additionally, desorption at the initial stage

of soil immersed in water can cause increase of arsenic species concentration in solution. In the adsorptive dynamics during 0–24 h, only As(III) in 0–16 h and As(V) in 1–24 h of baiho soil were of the 3rd order, and the remaining arsenic species adsorbability of reaction system was followed 0 order reaction. Thus, this study will be helpful for further application in the treatment of arsenic contaminated Taiwan soils.

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