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Study on arsenic removal rate of combined process of coke-bed trickling filter and sedimentation

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Abstract: Arsenic contamination in groundwater has been recognized as a serious issue for health and environment because arsenic is a hazardous and toxic element; therefore, developing new technology is significant to remove arsenic from groundwater. Technology of arsenic removal has been in place through research to mitigate this problem, including aeration approach, Haix absorbent, precipitation, absorption process, and membrane technology. This study aims to evaluate and investigate new method of removing arsenic by applying the combined process of coke-bed trickling filter and sedimentation. One tubewell water from Koh Thom district, Kandal province, in Cambodia was chosen for the study. The concentration of total arsenic (As-T), arsenite-arsenic (As(III)), arsenate-arsenic (As(V)), pH, Mn, and Fe were analyzed. The combined process of coke-bed trickling filter and sedimentation was conducted with four different Phases (A, B, C, D). Phase A represents the flow rate of 1.0 L/day of pumping influent groundwater sample with aeration, and without introduction of iron. Phase B represents the flow rate of 1.0 L/day of influent sample with aeration and introduction of 25 mg/l of iron added. Phase C and D represents the flow rate 1.5 L/day and 2 L/day; respectively, of influent sample with aeration and introduction of 25 mg/l of iron added. The concentration of As-T was removed in Phase A by 29%. Whereas the concentration of As-T showed high effectively removal from 80 to 86% in Phase B, C and D. Specifically, the total concentration of As decreased from 356.25 μg/L to 48.75 μg/L, 108.75 to 21.625 μg/L, and 472.5 to 75 μg/L for Phase B, C, and D; respectively. In conclusion, this combined process could be an effective technique to remove As from groundwater.

Keywords: groundwater; sedimentation; coke-bed trickling filter; Total arsenic

1. INTRODUCTION

Arsenic (As) is one of the natural existence hazardouse element in groundwater. Water contamined with As has been concerned in global because As is metalloid group, unnoticeable, colorness, no flavor and aroma, also extremely toxicity in the environment (Anjum et al., 2009; Hernández-flores et al., 2018; Hu et al., 2014). More than 70 countries worldwide have been influenced by As pollution in groundwater including Bangladesh, India, China, Vietnam and Cambodia (Kang, 2016). Furthermore, the contamination of As in groundwater is very harmful to human health since As is mostly present as inorganic type (Bilal et al., 2018). In addition, the danger of inorganic arsenic species in groundwater is arsenite (As(III)), which is more toxicity than oxidized form (As(V)) due to its solubility (Nicomel et al., 2015; He et al., 2018). As a result, drinking water contaminated with As can cause to skin cancers, neurological effects, cardio-vascular, cardiovascular, which caused to death (Agbaba et al., 2017; Omwene et al., 2019; Ravenscrof et al., 2011). Nowadays, arsenic contamination of tubewell in Cambodia has been recognized as a serious issue because of high level of arsenic concentration (Sok and Choup, 2017). Kandal province, located the southest of Cambodia, is one of areas that As is contaminated in tubewell at the highest concentration (RDIC, 2016), and people are living at risk of consumption water polluted with As. Thus, arsenic-free water is hugely needed to support rural people life and health, and protect them from As toxicity. The As treatment has been studied to increase the number of tubewells in rural area to secure the As contaminated groundwater by applying cost-effective technology.

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Various accessible methods for As removal from groundwater have been developed such as ion exchange, oxidation, co-precipitation, lime treatment, adsorption onto coagulated flocs (Ahmed, 2015), selective-membrane methods by reverse osmosis (Sahu et al., 2018), adsorption onto metal oxides, and coagulation with iron (Karcher et al., 1999). The researchers have been trying to elaborate methodology and means for As removal from contaminated groundwater in economic way. Some developing countries prefer to operate an adsorption method which is easier in term of technology and affordable cost. Moreover, some materials including biochar, natural soil and metal oxides are supplied for adsorbent (Adlnasab et al., 2019). However, those developed methods are still not an optimum process to remove As; thus, with innovative research the process of coke-bed trickling filter and sedimentation is combined to study for As removal by controlling As loading rate. According to Endo (2018), coke has special characteristic in adsorption of As. Usual materials adsorbs arsenate-As (As(V)) in more high affinity than arsenite-As (As(III)). On the contrary, coke adsorbs As(III) and does not adsorb As(V), and provides hydrophobic and porous surface for microbial enrichment. This means coke surface provide habitat of arsenite oxidizing bacteria converting As(III) to As(V) that is the easier chemical form to be removed by many physical or chemical processes.

It is very important to develop a new treatment technology for small communities due to the increasing of people living in rural area (Anjum et al., 2009). Hence in this work, the developed As removal process is considered low cost and convenient operation.

This study aims to evaluate the As removal performance of groundwater by applying combined process of coke-bed trickling filter and sedimentation. Moreover, the best flow rate to be supplied in this combined process is investigated, and the arsenic removal performance of the experimental system is assessed. Three flow rates of groundwater sample are studied. Parameters analysis are pH, total arsenic (As-T), arsenate-arsenic (As(V)), arsenite-arsenic(As(III)), total ferrous and total manganese. In addition, only one tubewell was taken from Toul Svay village, Kampong Kong commune, Koh Thom district, Kandal province to process in this experiment.

2. METHODOLOGY

2.1 Sample collection

Groundwater samples were collected from Toul Svay village, Kampong Kong commune, Koh Thom district, Kandal province for this study. The tube well depth is 24 – 25m. Water samples were gotten directly from water pump

and stored in the lid-stoppered polyethylene bottles, size 25 L with air spaces and stayed room temperature. Samples were transported from Kandal province to laboratory by car. No acids were added in the water sample during storage.

2.2 Experimental design

Fig.1. presents the schema of installation for the As removal system. Groundwater was pumped to go through the coke-bed trickling filter by pumping machine. Coke-trickling filter beds have 8-centimeter in height, 6.5-centimeter in diameter and packed coke weight were 184.8g in average.

Fig.1. Schema of experiment design for As removal

2.3 Experimental Procedure

The combined process of coke-bed trickling filter and sedimentation tank was divided into two reactors. There are four Phases designed for this experiment (see **Table 1**), and aeration mixing was supplied for oxidation in raw groundwater. Since our natural groundwater contained less amount of iron, 25mg Fe (II)/ L is added (Huang et al., 2016; Ha et al., 2005), in Phase B, C&D. However, Phase A was controlling process without added any ferrous. Aerated groundwater was pumped with an Iwaki EHN-BVC11R Metering Pump (Iwaki Tokyo, Japan) to go up- flow through a coke-trickling filter bed. The flow rates of groundwater samples (in flow for the coke-bed trickling process) were kept as difference Phases model as shown in **Table 1**. Samples were collected at three points (M1, M2& M3) to

determine total As (T-As), arsenite (As(III)), arsenate (As(V)), pH, total iron and manganese.

Table 1 Phases of loading rate for arsenic removal experiment

Phases	Reactor 1	Reactor 2	Adding iron (25mg/L) in groundwater	Air aeration mixing
A	1.0L/ day	1.0 L/day	No	Yes
B	1.0 L/day	1.0 L/day	Yes	Yes
C	1.5 L/day	1.5 L/day	Yes	Yes
D	2.0 L/day	2.0 L/day	Yes	Yes

2.4 Water Sampling

Samples were collected from three different ports of the experiment apparatuses which are raw groundwater influent sample, coke filter effluent sample, and final effluent sample from sedimentation process. Thus, there are three samples in each reactor were taken to analyze parameters for this study.

2.5 Analytical methods

The pH value in influent groundwater sample, coke filter effluent sample and final effluent sample were measured by pH meter HM-30P (TOA DKK, Tokyo, Japan). For Fe (total) and Mn were analyzed according to Spectroquant Iron Test Kit 1.00796.0001 (Merck Millipore, Burlington, MA, USA) photometric method. The concentration of As-T and As(III) was measured by MQuant Arsenic test kit 1.17927.0001 (Merck Millipore, Burlington, MA, USA) which can detect As-T and As(III) by visual comparison of the reaction zone of the test strip with the fields of a color scale. The measuring range of determinations As: 0.005- 0.010- 0.025- 0.05- 0.10- 0.25- 0.50 mg/l As. Since the concentration of arsenic in groundwater was higher than 0.50 mg/l As, samples must be diluted three times with distilled water (20ml of each samples with 40 ml of distilled water). To determine the concentration of As(III), twenty milliliters of samples was filtered with an Arsenic Speciation Cartridge to remove pentavalent arsenic (As(V)), then diluted with distilled water same as measurement of As-T. The concentration of arsenate (As(V)) was determined as following formula.
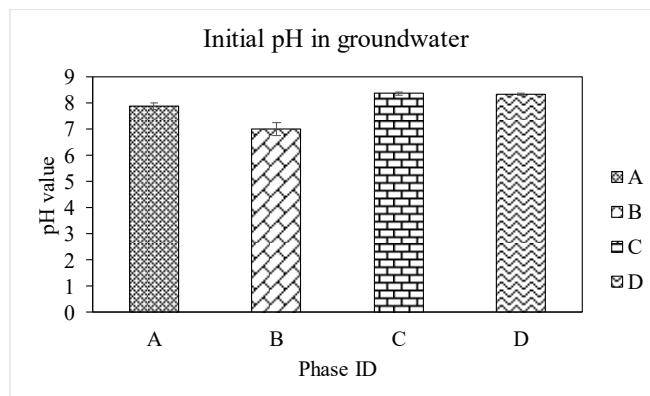
$$[As(V)] = [T\text{-}As] - [As(III)] \qquad (E.q\ 1)$$

3. RESULTS AND DISCUSSION

3.1 pH value

The pH is an important parameter to control the species of arsenic in groundwater.
Fig.2. showed the value of pH in raw groundwater with Phase A, B, C and D.

Fig.2. The pH value in groundwater for phase A, B, C and

Fig.2. illustrates the difference of pH value in groundwater in all Phases. In Phase B, the pH value is 6.99 ± 0.247 while pH value in Phase A, C and D are higher than neutral pH, which are 7.87 ± 0.123, 8.35 ± 0.0045, and 8.32 ± 0.033, respectively.
At pH value in Phase A and B, the predominance of arsenic species form as arsenate ($H_2AsO_4^-$ and $H_2AsO_4^{2-}$), and H_3AsO_3 as arsenite (Sato et al., 2002). The arsenite species (H_3AsO_3) is an uncharged electron which is difficult to remove from groundwater. Thus, the pre-oxidation state of arsenite is essential to form As (V). For pH value 8-9 in Phase C and D, arsenite had two forms with H_3AsO_3 and $H_2AsO_3^-$, and arsenate mostly formed as $H_2AsO_4^{2-}$ rather than $H_2AsO_4^-$ (Rajaković and Mitrović, 1992). Hence, Phase C and D arsenate and arsenite can be absorbed with iron and manganese on the sediment surface because of the electron charger form.

3.2 Concentration of Total Iron

Table 2 demonstrates the fluctuation of total iron (Fe-T) concentration in groundwater in four different Phases. In Phase A, it contained the lowest level of total iron. The concentration of ferrous iron (Fe(II)) in Phase A reached a bottom of horizontal axis, just 0.015 ± 0.01 because Phase A was not added any of iron into groundwater influent sample. The concentration of iron in groundwater sample was deficient. In Phase B, C, and D, twenty-five mg/l of Fe (II) was added into groundwater sample to make iron oxidization and precipitation with electron charge form. The concentration of iron in Phase B, C and D rose to 0.94 ± 0.11, 0.175 ± 0.04, and 0.42 ± 0.08, respectively.

The oxidation of iron is an essential state for the absorption of As(III) and As(V) species in groundwater sample tanks. The Fe(II) is oxidized to Fe(III) rapidly with oxygen dissolved (Roy et al., 2015). The adsorption of As(V) to amorphous iron oxide and goethite has also been found to be more beneficial than As(III) (Asere et al., 2019). In addition, Fe (II) almost oxidized to form of Fe (III) in air and precipitated at pH values 6.5-8.0, but As (III) did not oxidize to form As (V) at neutral pH. However, oxidation of As (III) occurred with the presence of Fe (III) (hydroxides) (Hug et al., 2001). When oxygen dissolves in water, ferrous oxidation reaction occures, the energy is released, and iron-oxidizing bacteria are able to live. This energy and bacteria in groundwater also affect the chemical reaction of arsenite oxidation, which increases oxidation rate of As(III). Hence, the low concentration of Fe(II) in Phase A leaded to slow oxidation of As(III).

Table 2. The different concentration of iron and manganese in groundwater in four phase

Phases	A	B	C	D
Fe	0.015	0.94	0.175	0.42
Mn	0.175	1.955	0.285	1.83

3.3 Concentration of Manganese

The concentration of total manganese (Mn-T) in groundwater in Phase A, B, C and D is illustrated in **Table 2** In phase B and D, it contained a high amount of Mn, but Phase A and C had a low amount of Mn. The concentration of Mn in Phase A was 0.175 ± 0.05, Phase B was 1.955 ± 0.28, Phase C was 0.285 ± 0.09 and Phase D was 1.83 ± 0.24.
Manganese in groundwater is also an essential factor in the combined process of coke-bed trickling filter and sedimentation to remove arsenic from groundwater. The oxidatized Mn coates surrounding soil grains and builds up adsorptive places (Luong et al., 2018). Nevertheless, the oxidation of Fe is faster than manganese. Compared to Fe, the rate of Mn oxidation is at least 106 times slower in water with a pH value close to neutral in Phase B. The oxidation of manganese accelerates with increasing pH value > 8. The chemistry of Mn is very complex, particularly in sediments and soil (Gounot, 1994). Therefore, it means Phase C and D which had pH value higher than 8, the oxidation of Mn increased and took more action than Fe.

3.4 Arsenic removal performance without Fe supplement in Phase A

The concentration of total arsenic (As-T), arsenite (As(III)) and arsenate (As(V)) of Grondwater influent sample (GW), Coke filter effluent sample (Co), and Final effluent (Fi) sample were demonstrated in **Fig.3(a)**. The concentration of As-T decreased from 723 µg/l in GW to 712.76 µg/l in Fi sample. Also, As(III) declined from 131.25 µg/l in GW to 38.90 µg/l in Fi sample. In contrast, As(V) increased from 591.75 µg/l in GW to 673.85 µg/l in Fi sample.
This results mean when groundwater sample went through coke-bed trickling filter, arsenite was absorbed on coke surface materials and oxidized to As(V), then As(V) was released to final effluent sample, which As(V) increased at final. Little amount of As(V) was precipitated with Fe(III) due to less concentration of iron in groundwater sample. As a result, the concentration of As-T remained high in Fi sample.

*3.5 Arsenic removal performance with Fe supplement in Phase B*

Fig.3(b). indicates various concentration of total arsenic, arsenite and arsenate in GW, Co and Fi sample. In period of processing in Phase B, concentration of As-T highly dropped down from 283.75 µg/l in GW to 131.875 µg/l in Fi sample. Furthermore, the concentration of As(III) declined from 55.92 µg/l in GW to 17.06 µg/l in Fi sample. Likewise, the concentration of As(V) decreased from 227.83 µg/l in GW to 94.81 µg/l in Fi.
The concentration of As-T decreased because As(III) oxidized to form As(V) by coke-bed trickling filter, and As(V) was absorbed with ferric iron (Fe(III)) form from Fe(II) oxidation, but As(V) is not easy to absorbed with Mn as Phase B had pH value near-neutral. With this pH, the oxidation of Mn is very slow (Gounot, 1994). Thus, the reduction in concentration of As(V) mostly was caused by precipitation process with iron oxidation.

3.6 Arsenic removal performance with Fe supplement in Phase C

The concentration of As-T, As(III) and As(V) in GW, Co and Fi sample is revealed in **Fig.4.2(c)**. The concentration of As-T went down from 86.75 µg/l in GW to 63.46 µg/l in Fi sample. The concentration of As(V) decreased from 84.25 µg/l in GW to 60.96 µg/l in Fi. However, the concentration of As(III) was slighly decrease from 7.5 µg/l in GW, and remained stable 2.5 µg/l in Co and Fi sample.
The low concentration of As(III) in GW showed that As(III) almost took oxidation place in GW by iron and manganese oxidation. As previous discussion when pH is higher than 8, the oxidation of Mn is increase, Mn (II) can be oxidized by
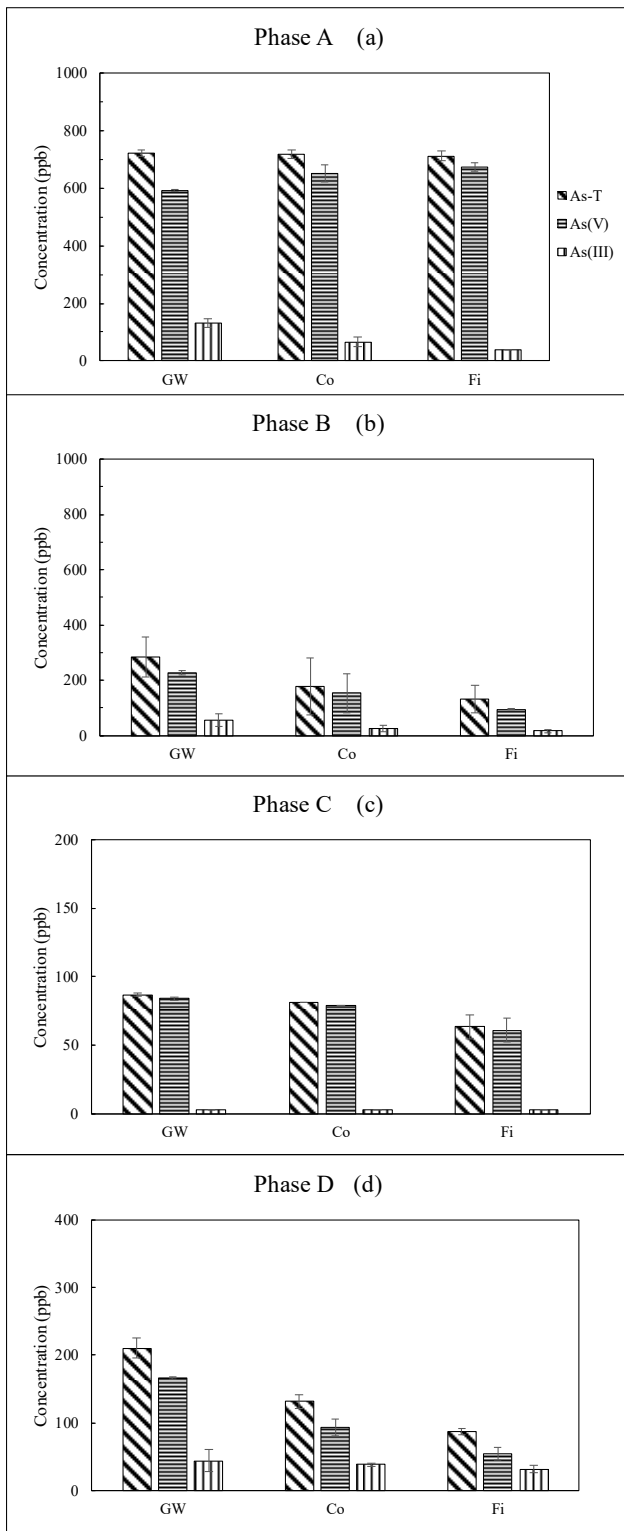
Fig.3. The concentration of As-T, As(V) and As(III) in influent groundwater (GW), coke-bed trickling filter effluent (Co), final effluent (Fi); Phase:A, 1l/day no added Fe(25mg/l); B, 1l/day added Fe(25mg/l); C, 1.5l/day added Fe(25mg/l); D 2l/day added Fe(25mg/l).

bacterium in groundwater and released energy (Majkić-dursun et al., 2014). Thus, manganese oxides as catalyst and oxidant increases arsenite oxidation rate (Tran, 2017). Hence, the concentration of As(V) is high in GW. However, it also has an effection of flow rate increasing as As(III) still remained in Fi sample.

3.7 Arsenic removal performance with Fe supplement in Phase D

Fig.3(d). shows the concentration of As-T, As(III) and As(V) in GW, Co and Fi sample. The concentration of As-T greatly declined from 210.56 µg/l in GW to 86.75 µg/l in Fi sample. Similarly, concentration of As(V) dropped from 166.56 µg/l in GW to 54.83 µg/l in Fi. Whereas the concentration of As(III) was slightly change in GW with 44 µg/l, Co with 38.5 µg/l and Fi with 31.92 µg/l.

The highly decrease of As-T is because of As (V) absorption with manganese and iron oxidation. Since Phase D had pH value with 8.32, manganes oxidation happened rapidly. The oxidatized Mn coates surrounding soil grains and builds up adsorptive places (Luong et al., 2018). Thus, the absorption of As(V) is also increase. However, the effluent water still contained high concentration of As(III). It would be because of the increasing influent water flow rate in Phase D (2L/day). When the flow rate was increased, the resident time of coke-bed trickling filter with influent water was decreased which leaded to low As(III) oxidation by coke materials.

3.9 The percentage of arsenic removal

According to the data showed in **Fig.4.,** the different percentage of arsenic removal was obtained in Phase A, B, C, and D. The combined process of coke-bed trickling filter and sedimentation is effective to remove arsenic from groundwater by adding 25mg/l of iron (Fe(II)) in Phase B, C and D, as Fe(II) oxidation to form Fe (III) is an important stage to adsorb As(V) after As(III) oxidation to As (V) (Asere et al., 2019).
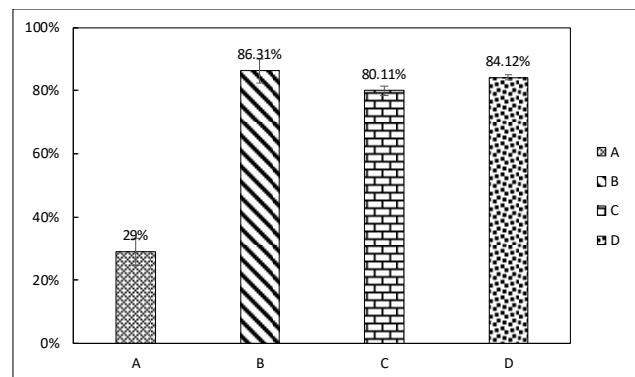
Fig.4. The percentage of total arsenic removal efficiency in GW, Co and Fi in Phase A, B, C and D

For Phase B, arsenic was removed with 86.31%, whereas Phase A arsenic could be removed only 29% of As.

The reason would be because Phase B contained high concentration of Fe and Mn in groundwater and acceptable pH value; therefore, arsenic concentration would be well removed.

The presence of Fe and Mn in groundwater demonstrated the beneficial precipitation and adsorption rate with As(V) (Gounot, 1994). In addition, the pH value in Phase B was 7 which As(III) mostly formed as unchangeable, so As(III) oxidation mostly occurred via coke-bed trickling filter. Moreover, Phase B had a low flow rate 1.0l/day, which means the resident time for arsenite (As(III)) oxidation by coke surface material may be longer than Phase C and D. Thus, As(III) was completely oxidized to As(V) during experiment processing in Phase B.

4. CONCLUSIONS

The results obtained from this study showed the efficiency of arsenic removal performance in the combined process of coke-bed trickling filter and sedimentation. This combining process revealed that arsenic removal rate could reach 80% to 86%. Moreover, the flow rate could be significant factor for resident time of As(III) oxidation with coke surface material. Thus, the flow rate 1.0L/day in Phase B would be a good condition flow rate for sample influent to supply in the process of added 25mg/L iron in groundwater sample with pH value near neutral.

Further study, the pH value in groundwater sample is very important to control changing water quality. Thus, pH value should be controlled.

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