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Development of a novel process for arsenic(V) removal from water using Fe-ettringite

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ABSTRACT

The Fe-ettringite formation and its arsenic removal capacity were investigated. The synergetic effect of initially injected ferric iron and formation of ettringite crystal in later stages contributed to improved removal capacity. The As(V) removal capacity by the formation of As(V)-containing Fe-ettringite (As/Fe-ettringite) was estimated through kinetic and equilibrium batch experiments. The As/Fe-ettringite formation was induced by injecting CaO and $FeCl_3$ into synthetic As(V) wastewater (1.33 mM). Different molar ratios of As(V)/Ca/Fe (1:2:1, 1:4:2, 1:6:3, 1:8:4, and 1:10:5) were experimented in batches, the ratio of 1:6:3 showed the best removal efficiency (≈95%) among them. The control experiments in which CaO (8.008 mM) and $FeCl_3$ (4.004 mM) were used individually showed much lower arsenic removal capacities (2.5% for CaO and 32.5% for $FeCl_3$) compared with the combined system of CaO and $FeCl_3$ which led to As/Fe-ettringite formation. Equilibrium experiments were conducted in the pH range from 3 to 13, and the result showed As(V) removal by As/Fe-ettringite formation was significant in the pH range from 10.9 to 11.8, which was favorable for the formation of ettringite phases. Well-crystalized As/Fe-ettringite was observed in the FE-SEM micrographs of samples, and the needle-like crystals were most abundant and clear in the sample taken at pH of 12.

Keywords: Ettringite; Arsenic; Precipitation; Ferric iron; Calcium

1. Introduction

Ettringite-containing aluminum ($Ca_6Al_2O_6(SO_4)_3{\cdot}32H_2O$, Al-ettringite) commonly occurs in hydrated cementitious materials, generated by the hydration reaction of calcium, alumina, and sulfate. Al-ettringite is the most common in hydrated cement and is usually referred to as the AFt (Al_2O_3-Fe_2O_3-tri) phase. The exchange capacity of ettringite especially related to oxyanion contaminants, such as AsO_4^{3-}, CrO_4^{2-}, and SeO_4^{2-} has been an area of interest for many researchers, and it has been investigated in terms of waste stabilization using cementitious materials [1–5]. It is well known that Al-ettringite commonly plays an important role in the

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immobilization of contaminants in cement-based solidification/stabilization by adsorption of the contaminants into the intercolumn channels and by the replacement of sulfate [6–8]. Furthermore, the aluminum site in Al-ettringite can be readily substituted with other trivalent cations, depending on the type of replacement of trivalent cation and divalent anion that are reported during different ettringite phases [1,9].

When ettringite is used for the treatment of contaminant, the contaminant removal mechanism may take place in two different ways. The contaminant could get adsorbed in the interchannels of already formed ettringite structure. Another way is that if the contaminant is anionic, then during the formation of ettringite by coprecipitation the contaminant could get incorporated into ettringite crystal. The second mechanism is more promising, because the amount of arsenate uptake in the coprecipitation experiment was more efficient for contaminant removal than the first mechanism, due to the greater accessibility of interchannel sites during coprecipitation [6]. The simultaneous removal of As(V) during the formation of ettringite can be a more effective method for removing As(V) from aqueous solution. Several researchers have developed wastewater treatment technologies using the principle of ettringite formation/coprecipitation. Approximately, 90% of sulfate in the acidic wastewater from an aluminum anodizing industry was removed by the addition of calcium oxide and calcium aluminate with molar ratios of $Ca^{2+}/Al^{3+}/SO_4^{2-}$ of ettringite [10]. The removal of arsenic and sulfate from acid mine drainage was effectively achieved by including the precipitation of ettringite with the addition of fly ash (substitute for lime in the SAVMIN process), gypsum, and amorphous $Al(OH)_3$ [11].

In this study, Fe-ettringite was chosen to remove As (V) in an aqueous phase. Fe-ettringite also occurs in cement hydrate phases, although it is less prevalent than Al-ettringite [12,13]. Fe-ettringite was reported to behave similarly to Al-ettringite and to be more stable in highly alkaline environment [14]. In this alkaline environment, the other heavy metals that coexist in the arsenic-contaminated wastewater can also be effectively and simultaneously eliminated by the formation of hydroxide precipitates. These advantages can make the operation and management of a new treatment method using Fe-ettringite easier and more cost-effective.

The objective of this study was to develop a new process for the removal of arsenic from wastewater by precipitation in the form of As(V)-containing Fe-ettringite (As/Fe-ttringite). Conventionally, ferric iron has been widely used for arsenic removal as a coagulant for coprecipitation due to its high affinity for arsenic as well as its cost-effectiveness [15–18]. Therefore, the synergetic effect from the combination of both the ferric iron that was injected initially and the formation of the As/Fe-ettringite crystal in later stage was expected to noticeably improve the arsenic removal capacity. In order to achieve this goal, various molar ratios of CaO and $FeCl_3$ with respect to arsenate concentration were examined to investigate the As(V) removal efficiency by the formation of As/Fe-ettringite. Then, the amount of As(V) uptake in the artificial wastewater was tested in different pH ranges, and FE-SEM analysis was performed to confirm the formation of precipitate of the As/Fe-ettringite.

2. Materials and methods

2.1. Materials

Synthetic wastewater containing 100 mg/l (1.33 mM) of arsenate was made using sodium arsenate (Na_2HAsO_4 $7H_2O$, Yakuri Pure Chemicals Co., Ltd.) dissolved in ultrapure water. One liter glass bottle was used as a reactor, and the polyethylene lid for the reactor was designed to plug the agitator arm of jar tester apparatus, pH meter, and to prevent CO_2 contamination (Fig. 1). The pH meter was placed in the system to monitor changes in the pH of the solution. In order to induce the formation of As/Fe-ettringite, the designated amounts of calcium oxide (CaO, Showa Chemicals Inc., Japan) and ferric chloride ($FeCl_3$, Showa Chemicals Inc., Japan) were injected into As(V)-containing water and then mixed using a jar tester (WiseStir™ Jar Tester; DH.CV006010). HCl (35%,

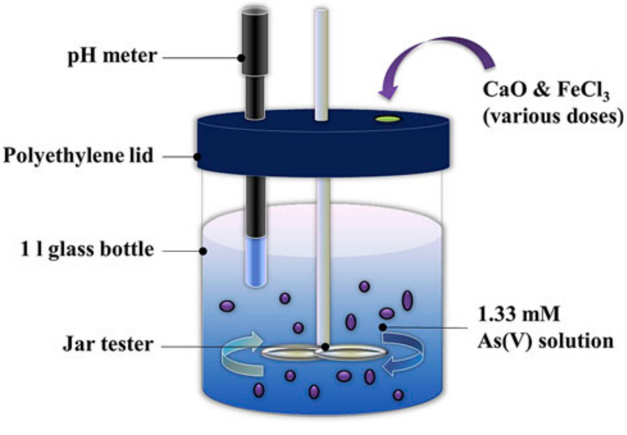

Fig. 1. The schematic diagram of the reactor for the experiment on the removal of As(V) by As/Fe-ettringite formation.

Matsuneon Chemicals Ltd.) and KOH (Duksan Pure Chemical Co., Ltd.) solutions were used to adjust the pH level of system.

2.2. Experimental methods

2.2.1. Effect of the component ratios of As(V) and cations on As(V) removal

Different sets of artificial As(V)-containing water were prepared with various doses of CaO and $FeCl_3$ to observe the effect of the molar ratios of cations (calcium and ferric iron) on the formation of As/Fe-ettringite. The stoichiometric ratio of As(V)/Ca/Fe was calculated to be 3:6:2 using the formula for As/Fe-ettringite ($Ca_6Fe_2(HAsO_4)_3{\cdot}xH_2O$) that is analogous formula with that of generic Al-ettringite ($Ca_6Al_2(SO_4)_3{\cdot}26H_2O$). However, we thought that the more ferric iron had to be added due to the possibility of the coexistence of different arsenate species in the form of $HAsO_4^{2-}$ and AsO_4^{3-} within the pH range (11–12) used in this experiment. Therefore, the basic stoichiometric ratio of As(V)/Ca/Fe for promoting the formation of As/Fe-ettringite was determined to be 3:6:3 (=1:2:1). Based on the aforementioned basic ratio, CaO and $FeCl_3$ were injected into the following different molar ratios of As(V)/Ca/Fe as 1:2:1, 1:4:2, 1:6:3, 1:8:4, and 1:10:5, in individual sets (Table 1). Rapid stirring for 5 min at 100 rpm and slow stirring for 60 min at 40 rpm were carried out to enhance the formation of As/Fe-ettringite crystal using the jar test in a closed reactor. After stirring, the contents of the reactors were allowed to settle for 20 min. After settling, the supernatant solution was collected and filtered through 0.45-μm Whatman® syringe filter. Samples of the filtrate were taken and analyzed in duplicate for the residual As(V) concentration.

Table 1
Labels, molar ratios, and injected concentrations of the individual sets in the experiment to investigate the effects of the component ratios on As(V) removal by the formation of As/Fe-ettringite

Labels	Molar ratios (As(V)/Ca/Fe)	Injected concentrations (mM)		
		As(V)	Ca	Fe
Set I	1:2:1	1.335	2.669	1.335
Set II	1:4:2	1.335	5.339	2.669
Set III	1:6:3	1.335	8.008	4.004
Set IV	1:8:4	1.335	10.678	5.339
Set V	1:10:5	1.335	13.347	6.674

2.2.2. Effect of solution pH on As(V) removal

Ettringite is known as a mineral product of cement hydration, and its crystal stability is significantly affected by pH conditions [5,11,19]. The pH level also plays a very important role in arsenic removal [6]. Therefore, the experiments were designed to investigate the effects of the pH level on the formation of As/Fe-ettringite crystals and to check the As(V) removal efficiency. The injection doses of CaO and $FeCl_3$ were determined to be 8.008 and 4.004 mM (set III in Table 1), respectively, based on the previous As(V) removal experiments (section 2.2.1); 1 M HCl and 5 M KOH were used to control the solution pH. The pH values in the individual sets were adjusted to 3, 5, 7, 9, 11, 12, and 13. The experiment was conducted using the same methods as in the previous experiments (section 2.2.1), and the kinetics of the As(V) removal were measured.

2.3. Analytical methods

The concentration of arsenate was analyzed using an atomic adsorption spectrophotometer (AAS, Vario 6, Analytikjena) connected to hydride AsH_3 (arsine) generator (HS-55, Analytikjena) at a wave length of 193.7 nm. As(V) samples were prepared as duplicate, and they were pretreated by reducing As(V) to As(III) before being introduced into the measuring instrument. The average value from the duplicate analyses was reported in this study. As(V) removal efficiency was calculated by the following expression:

$$\text{As(V) removal efficiency}\,(\%) = (C_0 - C)/C_0 \times 100 \qquad (1)$$

where C_0 and C are the As(V) concentrations of the synthetic wastewater sample and a filtrate from As(V) removal experiments, respectively.

SEM analysis was carried out to observe the surface morphology of As/Fe-ettringite. The precipitates were dried for 3 days in a vacuum desiccator and were analyzed using Carl Zeiss SUPRA 55VP field-emission scanning electron microscope (FE-SEM) with sputter coating of platinum powder (BAL-TEC/SCD 005). All analyses were conducted at room temperature (23 ± 2°C) under isocratic conditions.

3. Results and discussion

3.1. Effect of the component ratios of Fe-ettringite

The arsenate removal efficiency of the formation of As/Fe-ettringite with different molar ratios of As(V)/Ca/Fe was examined. Fig. 2 shows the changes of arsenate uptake along with the molar ratios of calcium

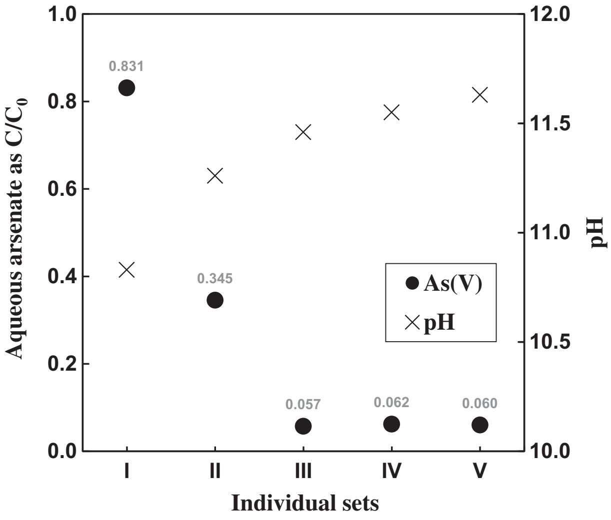

Fig. 2. Aqueous arsenate as C/C_0 with individual sets with different molar ratios of As(V)/Ca/Fe for the formation of As/Fe-ettringite. Initial concentration of arsenate was 1.33 mM.

and ferric iron in the As(V)-contaminated water. The results showed that the As(V) removal efficiency increased with an increase in the amount of cations injected into sets I (1:2:1), II (1:4:2), and III (1:6:3). In particular, 94.3 mg/l of the arsenate ($C/C_0 = 0.057$) was adsorbed into set III, when approximately 8 mM of calcium oxide and 4 mM of ferric chloride were injected, respectively. The solution pH also increased with the increase in amount of cations injected, which occurred at a pH level of approximately 11.5 in set III. The increase in the arsenate removal was not significant in IV ($C/C_0 = 0.062$) and V ($C/C_0 = 0.060$) sets above a certain dosage of calcium oxide and ferric chloride. This result suggests that the injected molar concentrations of calcium and ferric should be at least 6 and 3 times more, respectively, than that of arsenate for the formation of As/Fe-ettringite. It is known that arsenic removal by coprecipitation (involving flocculation and coagulation) with ferric and/or aluminum salt increases with an increase in the acidity (pH), because the surface charge of iron oxide becomes more positive with a decrease in pH in order to provide favorable conditions for the sorption of anions [16,17]. The effective pH range for As(V) removal is considered to be between 5 and 8 for ferric ions [20]. In the present study, arsenate was effectively removed even at an alkaline pH. This result showed that the mechanism of arsenate uptake by the formation of As/Fe-ettringite was fundamentally different from that of coagulation/coprecipitation by the addition of ferric salt. The solubility of the synthesized Fe-ettringite was studied by aging of the hydrated mixture of $Fe_2(SO_4)_3·5.3H_2O$, CaO, and KOH in a CO_2-free atmosphere [14]. The solubility was measured in a series of precipitation and dissolution experiments at 20˚C and at pH values between 11.0 and 14.0, so that the solubility product (log $K = -44.7 \pm 0.7$) of Fe-ettringite had already emerged. The value appears to be analogous with that of Al-ettringite (log $K = -44.9 \pm 0.7$ at 25˚C), because of the resemblance of molecular structure, but it is much smaller than the solubility products of calcium arsenate [21] and ferric arsenate minerals [22]. The As/Fe-ettringite, in this study, was analogized to have a very low solubility, such as the ettringites in the aforementioned studies. Consequently, the significant removal of As (V) in alkaline pH levels can possibly be explained by the coprecipitation of arsenate as the form of As/Fe-ettringite.

On the other hand, there is a possibility that the arsenate uptake might have been induced by the hydroxide precipitation of calcium and ferric iron [11,17,18]. In order to elucidate the As(V) removal capacity of Fe-ettringite, the control experiments using CaO (8.008 mM) and $FeCl_3$ (4.004 mM) were performed independently and their capacities were compared with that of As/Fe-ettringite (Fig. 3). In the case of As/Fe-ettringite, it was observed that the arsenate concentration rapidly decreased from an initial value of 1.33–0.06 mM in 5 min, and then, it remained at a constant concentration. However, relatively low arsenate removal efficiencies were observed with only CaO and $FeCl_3$. Merely 0.08 mM of arsenate removal was observed in 8.008 mM of CaO throughout the experiment, while approximately 0.467 mM of arsenate was removed in 4.004 mM of $FeCl_3$. These results indi-

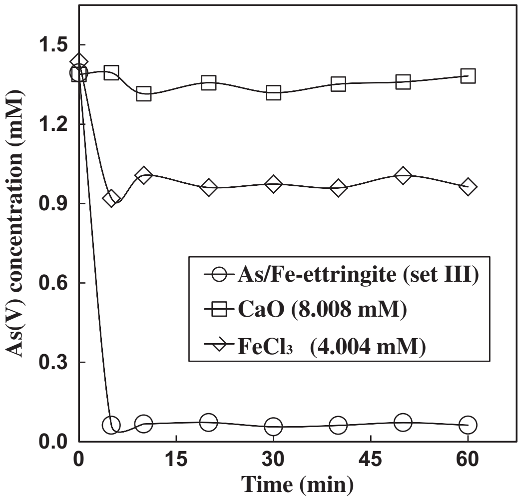

Fig. 3. Variations of residual arsenate concentrations of As/Fe-ettringite (set III), CaO (8.008 mM), and $FeCl_3$ (4.004 mM).

cate that individual sets of $FeCl_3$ and CaO could not achieve the same As(V) removal capacity of As/Fe-ettringite, which supports that there is the synergetic effect from the combined system of CaO and $FeCl_3$ which led to the formation of As/Fe-ettringite.

Almost 100% arsenic removal can be achieved by the coagulation process using ferric iron as the sole coagulant that leads to the formation of highly insoluble ferric arsenate (e.g. $FeAsO_4$). However, still there are some problems associated with this type of treatment such as consumption of an excess amount of reagent and the production of excess precipitates [23]. Therefore, this coagulation process is seldom applicable in the field. Previous studies showed up to 98.1% arsenic removal, when the As(V) to ferric chloride injection ratio was 1:60 (w/w) in synthetic contaminated water [18]. In contrast, the results of the present study indicate that treatment using the precipitation of As/Fe-ettringite can be regarded as a more economical and eco-friendly application, because relatively small amounts of cations are needed [As(V)/Ca/Fe ≈ 3:6:2 (w/w)] to achieve approximately 95% As(V) removal.

3.2. Effect of pH

The equilibrium experiments were performed under various solution pH ranges to estimate the effect that pH levels have on arsenate removal by the formation of Fe-ettringite. In consideration of the result of the previous experiment (section 3.1), the amount of calcium and ferric injected of set III (As(V)/Ca/Fe = 1:6:3) was used in the equilibrium experiments. Fig. 4 shows the residual As(V) concentrations by formation of As/Fe-ettringite along with various pH ranges. It was observed that the residual As(V) concentrations in the supernatant were different at different pH ranges. The arsenate uptake was predominant at pH levels of 2.5 and between 11 and 12. The concentration of the residual As(V) in the solution was reduced to 0.046 mM at a pH level of 2.6. Lower pH levels are more effective for the treatment of arsenic-contaminated water by iron salt, because coprecipitation is markedly enhanced by both the arsenate species (e.g. $H_2AsO_4^-$) and highly positively charged surface of the iron oxide with a decrease in the pH levels [22,24]. On the contrary, the arsenate uptake rapidly decreased with an increase in the pH range to between 3 and 9. This result implies that the cations can barely be crystallized to form As/Fe-ettringite in a neutral environment, because the formation of ettringite was predominant and stable in the pH range of 11.5–12.5 [6,25]. On the other hand, noticeable changes in the arsenate removal were observed at pH values of approximately 5 and 7. The residual As(V) concentrations were determined to be 0.29 mM at a pH of 4.9 and 0.319 mM at a pH of 7.1, compared to the mean concentration which was 0.375 mM in the pH range of 3–9. Scorodite ($FeAsO_4 \cdot 2H_2O$) and $Fe_3(AsO_4)_2$ can be formed in these pH ranges [22]. Therefore, the coprecipitation of the ferric iron minerals might have occurred through the formation of such minerals accompanied by the formation of a little As/Fe-ettringite. Meanwhile, the

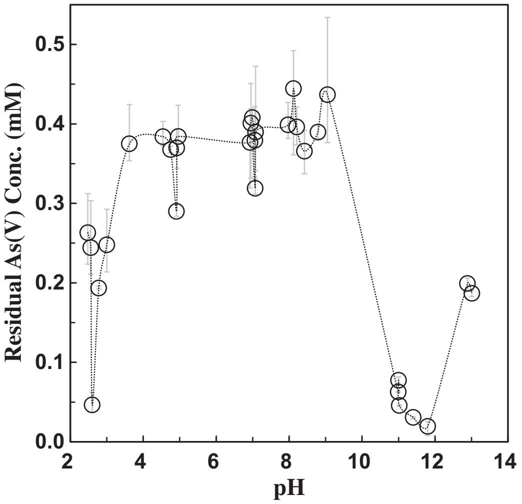

Fig. 4. Residual arsenate concentrations along with various pH ranges and their effects on As(V) removal by As/Fe-ettringite formation (set III). Initial As(V) concentration was 1.33 mM.

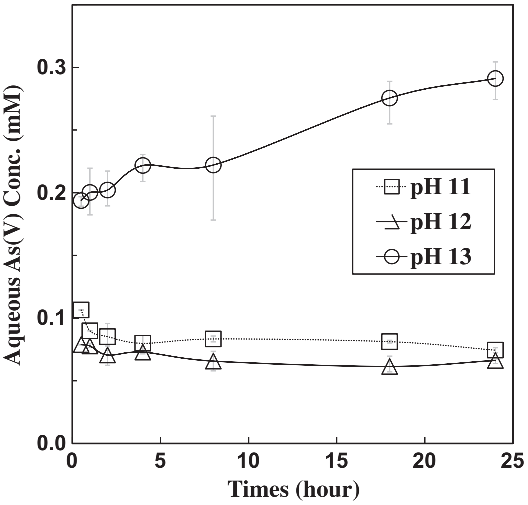

Fig. 5. Changes in the arsenate concentration in alkaline pHs by As/Fe-ettringite formation after 30 min of coprecipitation. Initial As(V) concentration was 1.33 mM.

experimental results of the present study in an alkaline pH agree with the results of previous studies that reported the favorable pH conditions for formation of ettringite phase are between 11 and 12.5 [25–27]. The residual As(V) concentrations decreased significantly at a pH greater than 10.9, with more than 1.31 mM of arsenate being removed at a pH of 11.8. A pH range of 11.0–11.8 was obtained without any additional pH adjustment. Therefore, the As(V) removal process using As/Fe-ettringite can be considered as a highly applicable process. The arsenate uptake capacity was reduced around pH 13. This result could be because the ettringite phases are unstable above pH 12.5 [26,27], and the formation of portlandite ($Ca(OH)_2$) was predominant at this pH [10]. This depletes the available calcium and the negatively charged ferric hydroxide complex including $Fe(OH)_4^-$ that are essential for the formation of ettringite. Fig. 5 depicts the results of the kinetic test at fixed pH levels of 11, 12, and 13. The As(V) concentrations measured before 30 min were omitted to observe a trend in the arsenate release. The released arsenate concentration gradually increased from 0.194 to 0.291 mM at a pH of 13. On the contrary, the concentration was maintained throughout the experiment at both the pH levels of 11 and 12. This result supports the hypothesis that the formation of As/Fe-ettringite became unstable at a pH level greater than 12 due to the calcium consumption by the portlandite precipitation and/or the formation of negatively charged ferric hydroxide complexes.

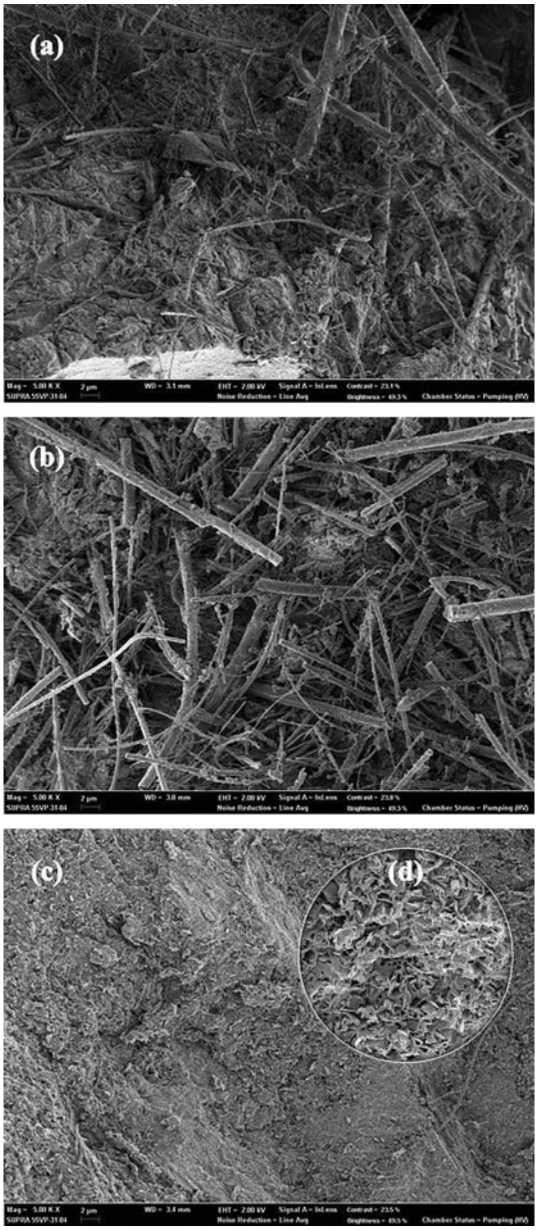

Fig. 6. FE-SEM images of As/Fe-ettringite formed at (a) pH 7 (×5,000), (b) pH 12 (×5,000), (c) pH 13 (×5,000), and (d) pH 13 (×20,000).

3.3. FE-SEM analysis

The evidence of formation of As/Fe-ettringite was confirmed by FE-SEM analysis, as shown in Fig. 6. The samples for FE-SEM analysis were collected from the arsenate removal experiments at pH levels of 7, 12, and 13. A needle-like crystal, which is a characteristic of ettringite, was observed in the samples at pH levels of 7 and 12. The more abundant and clear crystals were predominant in the micrograph of the samples at a pH level of 12, in comparison with those found at a pH level of 7, because an alkaline pH is the favorable environment for ettringite formation. However, the ettringite crystals were rarely found in the samples at a pH of 13. Instead, the plate-like forms of portlandite were predominant throughout the micrographs at a pH of 13. The results of the FE-SEM analysis are in agreement with the results of the batch experiments.

4. Conclusions

The As(V) removal capacity by the formation of As/Fe-ettringite was estimated in the present study. Several of the noticeable results and findings are summarized below: (1) Approximately, 95% efficiency of As(V) removal was achieved through the As/Fe-ettringite formation, when the molar ratio of As(V)/Ca/Fe was set to be 1:6:3. (2) The best removal efficiencies were found to be in the pH range of 10.9–11.8, which was the range that was found to favor the formation of the ettringite phases. The process using As/Fe-ettringite has an advantage, because relatively small amounts of reagents (approximately 1/23) are needed compared with the previous methods of arsenic removal by iron salt. In addition, in basic conditions (pH levels of 11–12) that are favorable for the As(V) removal reactions by the formation of As/Fe-ettringite, the other heavy metals that coexist in the arsenic contaminated wastewater can also be effectively and simultaneously eliminated by the formation of hydroxide precipitates. Favorable pH conditions for the simultaneous removal of heavy metals are achieved without pH adjustment.

Acknowledgments

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