

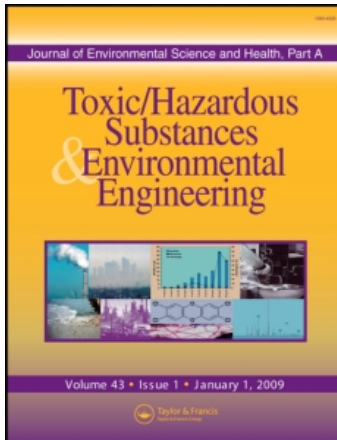
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## As(V) Removal from Aqueous Solutions by Coagulation with Liquid Phase of Red Mud

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### ABSTRACT

As(V) removal by using liquid phase of red mud (LPRM) is reported in this article. The experimental section includes characterization of LPRM, as well as As(V) removal from arsenical aqueous solution mixed with LPRM by coagulation in the column. As(V) removal study was divided into two parts; neutralization of LPRM-arsenical solution mixtures with acid solution accompanied with air-agitation and neutralization of those mixtures with CO<sub>2</sub> gas. Effect of LPRM/(As(V) solution) volumetric ratio on the removal of As(V) by co-precipitation arsenic together with aluminum present as aluminate in the LPRM were studied. Al/As(V) molar ratio values on the removal of As(V) is evaluated. Results show that As(V) was removed effectively by LPRM with a volumetric LPRM/(As(V) solution) ratio of 0.1 from an arsenical solution in the As(V) concentration of 20 mg dm<sup>-3</sup>. For an efficient removal, it was found to be required an Al/As(V) molar ratio of 6–8. The results suggest that it is advantageous to use a waste material of red mud liquid phase in the treatment of arsenical wastewater, possibly conjunction with red mud solids as adsorbent that its adsorption ability has been demonstrated earlier.

*Key Words:* As(V) removal; Coagulation; Red mud; Bayer process residue.

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## INTRODUCTION

Arsenic is a semi metallic element that has been classified among the priority pollutant for its toxicity. Arsenic minerals are not abundant in the earth's crust. However, it is naturally found as minor minerals such as arsenopyrite in the sulfide ores. Arsenic can be also found in coal. Metallurgical use and toxic effects of arsenic were documented by early Greek writers. Modern usage of arsenic and arsenical compounds include formulation of pesticides, herbicides and desiccants, decolorization of glass, preservation of woods, paint manufacturing, and the production of semiconductors.

Arsenic in water most often originates from geogenic sources, although anthropogenic arsenic pollution does occur. Traditional arsenic contaminated sites include areas of mining activities and smelters, because most of the acid mine drainages from sulphidic ores and flue dusts from smelters contains significant levels of arsenic. Additionally, agricultural use of arsenic compounds, metallurgical waste discharge and coal combustion by products cause arsenic pollution.<sup>[1]</sup>

Inorganic arsenic species in contaminated industrial sites exist in As(V) (arsenates), As(III) (arsenites or arsenic sulfides), As<sup>0</sup> (elemental arsenic) and As(-III) (arsines) forms. In water, the most common species of arsenic are arsenate, which is more prevalent in aerobic surface waters and arsenite, which is more likely to occur in anaerobic ground waters. In the pH range of 4 to 10, the predominant As(III) compound is neutral in charge, while the As(V) species are negatively charged. For this reason, arsenite is more mobile as it is less strongly adsorbed on most mineral surfaces than the negatively charged As(V) oxyanions. Removal efficiencies for As(III) are poor compared to removal As(V) by any of the technologies evaluated due to the negative charge.

Precipitation, ion exchange, reverse osmosis, electro dialysis and adsorption are possible techniques for arsenic removal. Co-precipitation of arsenate with iron or aluminum ions is recognized as overall the most effective and practical existing method of arsenic removal.<sup>[2,3]</sup>

In the alumina production, a waste, generally called red mud is formed as a result of processing bauxites by the worldwide practiced Bayer method. Red mud is disposed as a slurry of 200 to 350 g dm<sup>-3</sup> solids content from an aluminum plant. The solid phase consists of very fine particles. The liquid phase accompanying solids consists mainly of a weak sodium aluminate solution the nature of which is alkaline.<sup>[4,5]</sup>

In our previous studies,<sup>[6,7]</sup> arsenic removal by using the solid phase of red mud and acid activated red mud were investigated. In present study, arsenate removal characteristics of liquid phase of red mud by co-precipitation with aluminum hydroxide is explored.

## MATERIALS AND METHODS

### Materials and Characterization

The red mud used was obtained from the Etibank Aluminum Plant, Seydişehir-Konya, Turkey. Red mud slurry was taken from outlet of washing thickeners.



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The slurry having a liquid/solid ratio of about 2.5 was allowed to settle. Liquid phase was separated by decantation and filtered through a suction filter. The filtrate was named as Liquid Phase of Red Mud (LPRM), stored and used in the study.

LPRM was subjected to chemical analyses for Al, Na, alkalinity and carbonate. The following methods were utilized for analyses: Atomic absorption spectrophotometry for Al,<sup>[8]</sup> flame photometry for Na,<sup>[9]</sup> and titrimetry for alkalinity.<sup>[8]</sup> In order to determine carbonate content of liquid sample,  $\text{CO}_3^{2-}$  was precipitated as  $\text{BaCO}_3$  from an aliquot of liquid phase. The precipitate was decomposed in known amount of 0.1 N HCl. The excess hydrochloric acid was titrated by 0.1 N NaOH and finally the  $\text{CO}_3^{2-}$  content was calculated from the amount of consumed hydrochloric acid.<sup>[10]</sup>

As(V) stock solution of  $1000 \text{ mg dm}^{-3}$  was prepared from the salt of  $\text{Na}_3\text{AsO}_4 \cdot 7\text{H}_2\text{O}$  (Merck 6284). Experimental solutions were prepared from stock solution by diluting with distilled water.

HCl solutions, the concentration of which were 0.1 to 1.0 M were used in pH adjustments in the experiments carried out by air. HCl used in the experiments and other chemicals used in analyses were of analytical grade.  $\text{CO}_2$  used in the related experiments was supplied from a cylinder.

### Apparatus

Coagulation experiments were conducted by using a glass column (i.d = 5.5 cm, ht = 25 cm) with a glass porous plate (Por. no = 0) at the bottom. The system was equipped with a pH meter, an air pump (or a  $\text{CO}_2$  gas cylinder) and a gas rotameter. A schematic diagram of the system is given in Fig. 1.

### Experimental Procedure

Experiments were performed in two different methods. In the first method, 200 mL of solutions in various arsenic concentrations were placed in the column, various amounts of LPRM were added and then water saturated air was sent from the bottom of column by means of air pump to obtain a homogenous mixing. The volumetric air rate was set as  $500 \text{ cm}^3 \text{ min}^{-1}$  by a valve fitted onto the line. pH adjustments were done by adding acid solution from a burette placed over the column. At the end of the predetermined treatment period (15 min), air pump was stopped and the samples were taken. The samples were filtered and analyzed. Through these experiments, the effects of amount of LPRM and pH on the arsenic removal were determined.

In the second method, the  $\text{CO}_2$  gas was used both for neutralizing and mixing purposes. The  $\text{CO}_2$  gas in the volumetric rate of  $125\text{--}500 \text{ cm}^3 \text{ min}^{-1}$  was passed throughout the column containing the mixtures of arsenate solutions and LPRM in various ratios until the equilibrium pH was obtained.

Most of the experiments were performed in duplicate and the mean values were considered. The values obtained in parallel experiments were found to vary within  $\pm 5\%$ .

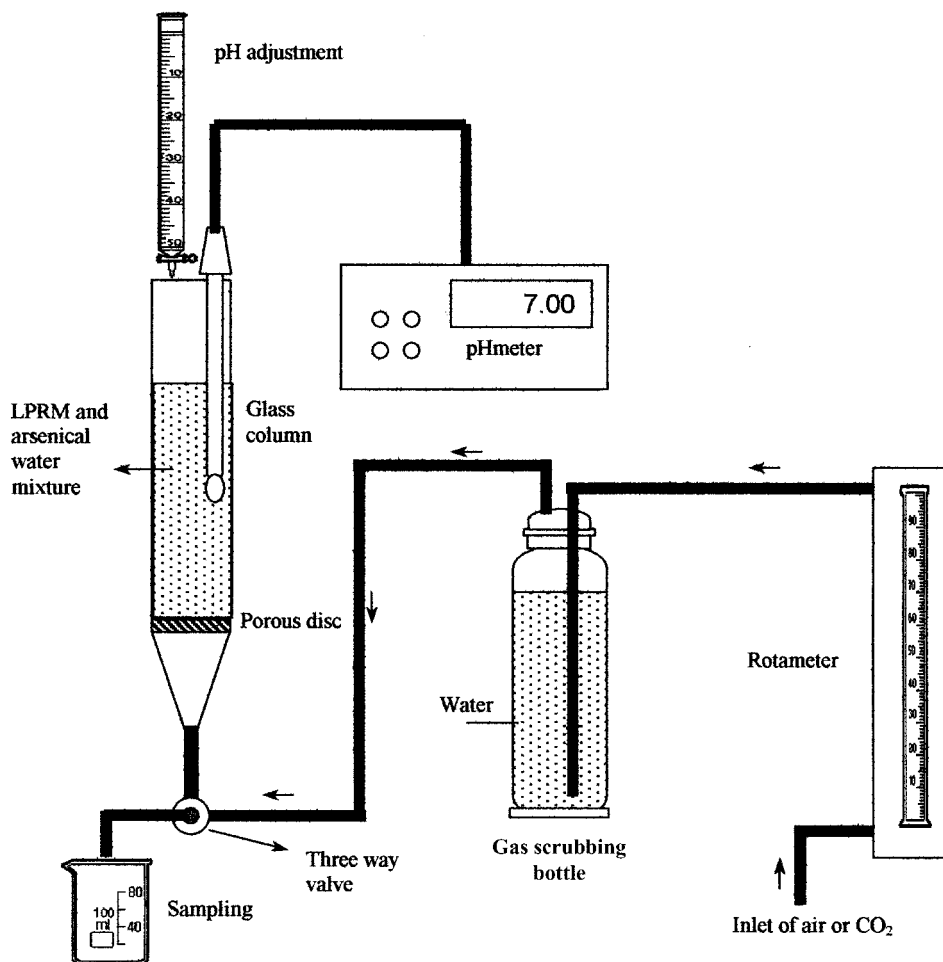


Figure 1. Schematic diagram of experimental apparatus.

### Methods of Analysis in Effluents

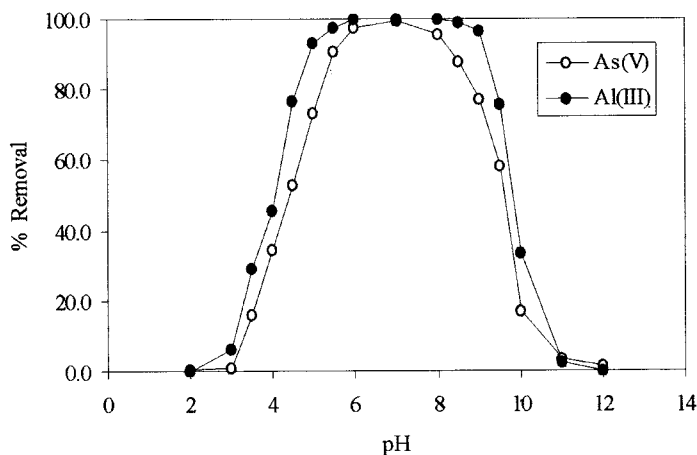
Following the filtration of the effluent solutions, arsenic was determined by using the silver diethyldithiocarbamate method.<sup>[9]</sup> Aluminum was determined by a Perkin-Elmer 370 atomic absorption spectrophotometer, whereas sodium was determined by an Eppendorf flame photometer. pH of solutions was measured by a Mettler Delta 350 pH meter which can save pH values acquired in narrow time intervals.

### RESULTS AND DISCUSSION

The composition of the liquid phase of red mud (LPRM) used in the study is given in Table 1. The LPRM contains significant amount of aluminum in the form of

**Table 1.** Analysis of liquid phase of red mud (LPRM).

pH	Total alkalinity (mg CaCO <sub>3</sub> dm <sup>-3</sup> )	Na (mg dm <sup>-3</sup> )	CO <sub>3</sub> <sup>2-</sup> (mg dm <sup>-3</sup> )	Al <sup>a</sup> (mg dm <sup>-3</sup> )
12.5	10850	2980	1720	620

<sup>a</sup>Al is in the form of aluminate.**Figure 2.** Effect of pH on the removal of As(V) by using HCl solution-air [As(V) solution volume: 200 cm<sup>3</sup>; Initial As(V) conc.: 50 mg dm<sup>-3</sup>; LPRM/(As(V) solution) ratio: 0.2; air flow rate: 500 cm<sup>3</sup> min<sup>-1</sup>; time: 15 min].

aluminate (AlO<sub>2</sub><sup>-</sup>). As seen from Table 1, the solution is highly alkaline. Upon neutralization, an aluminate based coagulant can form precipitates from arsenic contaminated aqueous system that cause arsenic removal.

To test the viability of the use of LPRM for the removal of arsenate from aqueous solution, both acid and CO<sub>2</sub> neutralization experiments were conducted.

In the first series of experiments, the mixture of LPRM and arsenic containing solution (50 mg-As(V) dm<sup>-3</sup>) in the ratio of 1/5 (v/v) was neutralized by acid solution accompanied with agitating by air. As(V) removal yield and precipitated aluminum percentage depending on pH are shown in Fig. 2. As seen from figure, efficient As(V) removal occurs in the close neutral pH range of 5.5–8 that aluminum precipitation follows similar pattern with arsenic removal. In this pH range, aluminum precipitates completely and more than 90% As(V) removal is achieved. Maximum arsenic removal (about 99.5%) is obtained at pH 7.

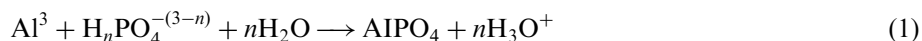
It is well known that aluminum has amphoteric character. For aluminum hydroxide, the minimum solubility pH can be calculated as 5.82 from the solubility product ( $S_{\text{pAl(OH)}_3} = 1.99 \times 10^{-33}$ ) that the calculated equilibrium concentration of aluminum is quite low.

The mechanism governing with As(V) removal includes precipitation together with aluminum as co-precipitates. Additionally, arsenate oxyanions can be adsorbed

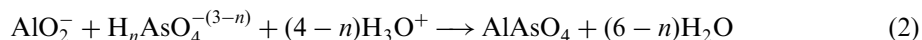


onto surface of formed aluminium hydroxide flocks. In this case,  $\text{pH}_{\text{zpc}}$  value of aluminum hydroxide and As(V) speciation become important factors. In the pH range of 6–7 which maximum As(V) removal was observed, it can be showed that the dominant species are  $\text{HAsO}_4^-$  and  $\text{H}_2\text{AsO}_4^{2-}$  by calculating from the pK values of  $\text{H}_3\text{AsO}_4$  ( $\text{pK}_{\text{a}1} = 2.19$ ;  $\text{pK}_{\text{a}2} = 6.94$ ;  $\text{pK}_{\text{a}3} = 11.5$ ). It has been reported that  $\text{pH}_{\text{zpc}}$  value of aluminum hydroxide is 5<sup>[11,12]</sup> which the surface of aluminum hydroxide flocks is charged positively below this pH value and it will be suitable for adsorption of negatively charged arsenate species. Since the maximum As(V) removal takes place at pH 7, it can be noted that the main removal mechanism cannot be focused on adsorption only.

On the other hand, the below reaction has been proposed for removing of ortho phosphates from aqueous solution by using aluminum salts as coagulation agent.<sup>[13]</sup>



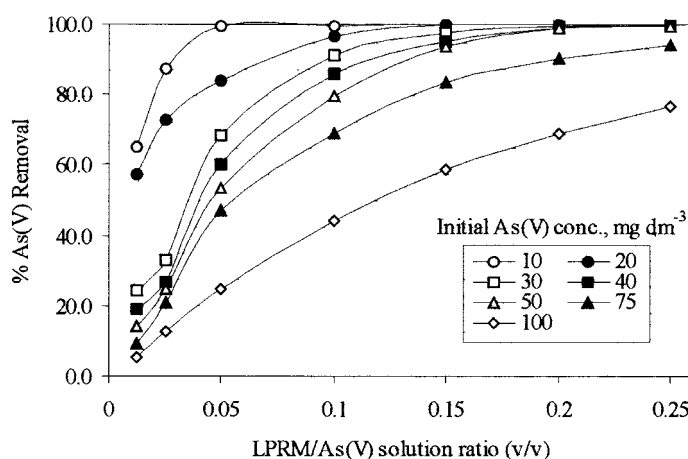
Phosphate ions are very similar chemically with As(V) since orthophosphoric acid have both similar structure and close  $\text{pK}_a$  values with arsenate acid. Thus, the similar reaction can be proposed for As(V) removal by using aluminate based LPRM as below:



However, beside reaction 2, following reaction does occur:



The effect of LPRM/(As(V) solution) volumetric ratio on the removal of As(V) from the solutions in various As(V) concentrations is shown in Fig. 3. For all As(V) concentrations, the removal efficiencies increase by LPRM/(As(V) solution) ratio.

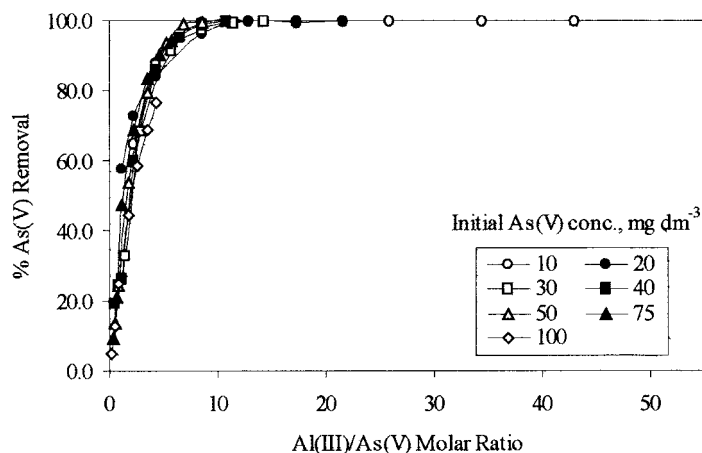


**Figure 3.** Effect of LPRM/As(V) solution ratio on the As(V) removal for various initial As(V) concentrations by using HCl solution-air [As(V) solution volume: 200 cm<sup>3</sup>; air flow rate: 500 cm<sup>3</sup> min<sup>-1</sup>; time: 15 min].



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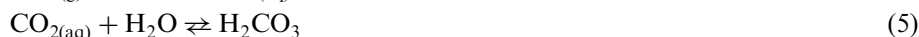


**Figure 4.** Effect of Al(III)/As(V) molar ratio on the As(V) removal for various initial As(V) concentrations by using HCl solution-air [As(V) solution volume: 200 cm<sup>3</sup>; air flow rate: 500 cm<sup>3</sup> min<sup>-1</sup>; time: 15 min].

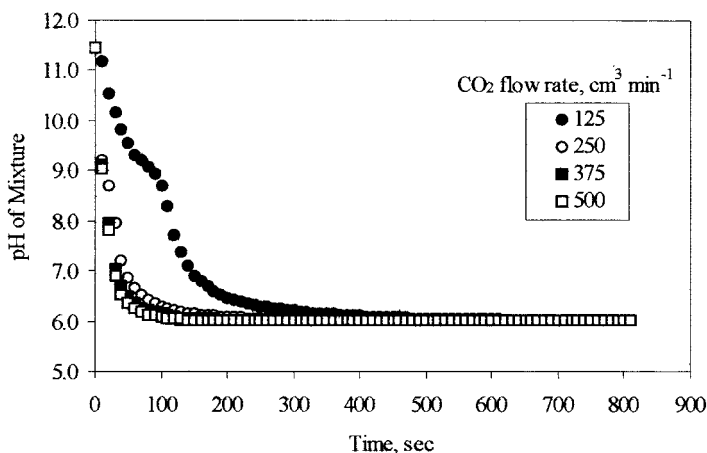
Variations of predetermined values of Al/As(V) molar ratio versus As(V) removals are illustrated in Fig. 4. As seen from the figure, As(V) removal depends strongly on the Al/As(V) molar ratio. Although required Al/As(V) molar ratio is 1 with respect to the stoichiometry of Eq. (2), the experimental molar ratio is found to be 6 to 8 for an efficient As(V) removal. This result indicates that aluminum hydroxide precipitates together with aluminum arsenate. As in the case of phosphate, it has been reported that the required aluminum compound dosage generally fall in the range of a 1 to 3 metal ion-phosphorus molar ratio.<sup>[13]</sup>

In the second part of the study, CO<sub>2</sub> gas was used as neutralization and agitating agent. Firstly, the mixture of LPRM and arsenic containing solution (50 mg-As(V)dm<sup>-3</sup>) in the ratio of 1/5 (v/v) was treated by CO<sub>2</sub> in various volumetric rates in order to determine the pH equilibration time. For this purpose, CO<sub>2</sub> gas in the various volumetric rates between 125 and 500 cm<sup>3</sup> min<sup>-1</sup> was passed through the solution in the column until the constant pH was obtained. The results of the variation of pH versus time are shown in Fig. 5.

As a mechanism, CO<sub>2</sub> gas dissolves under the formation of carbonic acid which neutralizes the alkalinity stemming from LPRM added. Flowing through the water, CO<sub>2</sub> reacts with water to an aqueous complex, CO<sub>2(aq)</sub>. Only a small part of the CO<sub>2(aq)</sub>, about 0.2–0.3%, react to form carbonic acid (H<sub>2</sub>CO<sub>3</sub>), which dissociates into hydrocarbonate (HCO<sub>3</sub><sup>-</sup>) and carbonate (CO<sub>3</sub><sup>2-</sup>). The absorption of CO<sub>2</sub> is part of the following equilibrium chain:







**Figure 5.** Effect of time and CO<sub>2</sub> flow rate on the pH of mixture [As(V) solution volume: 200 cm<sup>3</sup>; Initial As(V) conc.: 50 mg dm<sup>-3</sup>; LPRM/(As(V) solution) ratio: 0.2].

Theoretically, carbonic acid is a weak acid. But as the largest part of the dissolved CO<sub>2</sub> exists as hydrated CO<sub>2(aq)</sub> and not as H<sub>2</sub>CO<sub>3</sub>, its solution acts as a weak acid. Existing an alkaline component in the water, the equilibrium given in above reaction set is disturbed by the reaction of OH<sup>-</sup> and H<sub>3</sub>O<sup>+</sup> ions to form H<sub>2</sub>O. Tending towards the equilibrium, CO<sub>2</sub> dissolves again under the formation of hydrogen carbonate and carbonate. Taking into consideration the base fraction  $x_b = c_{\text{base}} / (c_{\text{acid}} + c_{\text{base}})$  and equilibrium constants of reactions, corresponding pH values in Fig. 6 can be calculated. In this connection, if the pH lies around 7, mostly HCO<sub>3</sub><sup>-</sup> is formed.<sup>[11,14]</sup>

As seen, pH of mixtures are equilibrated at around 6 for all volumetric rates studied (Fig. 5). However, this equilibration pH is reached at different times for various CO<sub>2</sub> gas rates. Required pH equilibration times for volumetric gas rates of 125, 250, 375 and 500 cm<sup>3</sup> min<sup>-1</sup> are 410, 190, 120 and 100 s, respectively. As expected, equilibration time decreases by increasing the volumetric gas rate. Also, it is obviously seen that neutralization process was accomplished in a short time.

The mixtures of LPRM and arsenic containing solutions in the various ratios (v/v) was subjected to CO<sub>2</sub> neutralization tests with a constant CO<sub>2</sub> volumetric rate of 500 cm<sup>3</sup> min<sup>-1</sup>. The results of the effects of LPRM/(As(V) solution) ratio (v/v) and Al/As(V) molar ratios on the removal of As(V) are given in Figs. 7 and 8, respectively. As can be seen, similar results are exhibited in experiments performed with CO<sub>2</sub> gas and acid solution as neutralizers. Finally, it can be said that CO<sub>2</sub> can be used as a cheap neutralizing and agitating agent.

In both neutralization processes carried out by using acid solution and CO<sub>2</sub> gas, it can be stated that 0.1 volumetric ratio of LPRM/(As(V) solution) is needed for an efficient removal from a solution in the initial arsenic concentration up to 20 mg dm<sup>-3</sup>. The higher the As(V) concentration is, the higher LPRM/(As(V)



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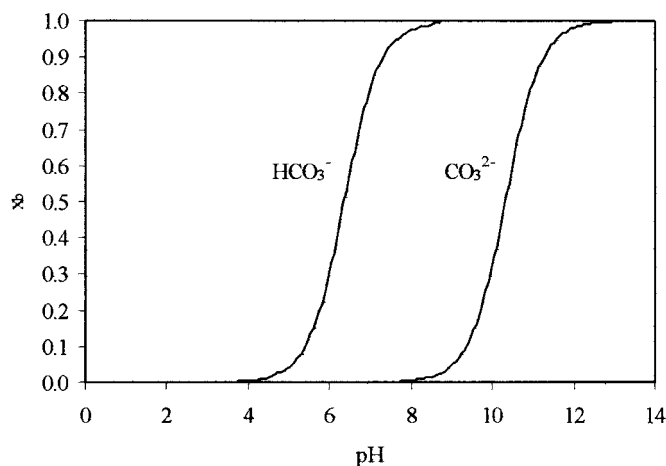


Figure 6. Dissociation equilibria of  $\text{CO}_2$ ,  $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$  vs. pH and base fraction  $x_b$ .

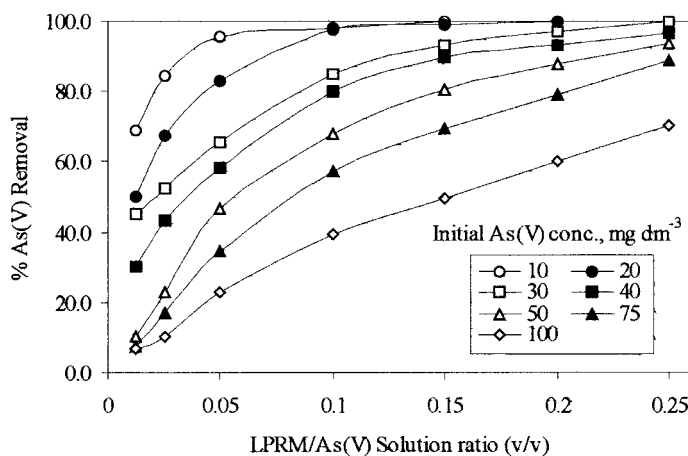
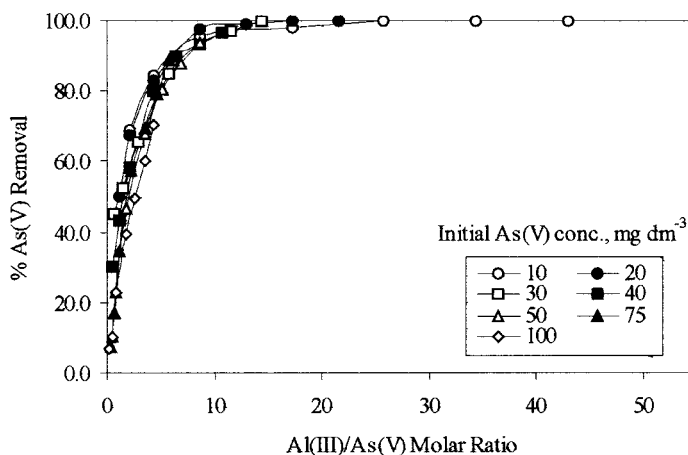


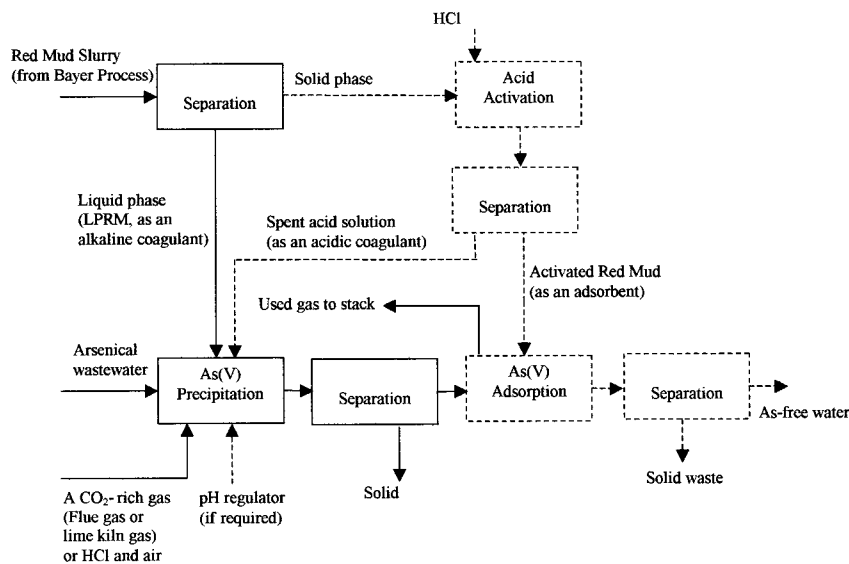
Figure 7. Effect of LPRM/(As(V) solution) ratio on the As(V) removal for various initial As(V) concentrations by using  $\text{CO}_2$  [As(V) solution volume:  $200 \text{ cm}^3$ ;  $\text{CO}_2$  flow rate:  $500 \text{ cm}^3 \text{ min}^{-1}$ ; time: 15 min].

solution) ratio is needed. For example, about 70% arsenic removal from a  $100 \text{ mg dm}^{-3}$  arsenical solution could be achieved with a ratio of 0.25.

These results indicate that this process could be utilized for an arsenic removal system as well as being developed for using a  $\text{CO}_2$ -rich flue gas or lime kiln gas substituting to pure  $\text{CO}_2$ . For a best result, however, solid phase of red mud consisted mainly of metal oxides may be recommended for completion of the removal process. In our previous studies,<sup>[6,7]</sup> it has been showed that red mud solids have significant arsenic adsorption capacity.



**Figure 8.** Effect of Al(III)/As(V) molar ratio on the As(V) removal for various initial concentrations by using CO<sub>2</sub> [As(V) solution volume: 200 cm<sup>3</sup>; CO<sub>2</sub> flow rate: 500 cm<sup>3</sup> min<sup>-1</sup>; time: 15 min].



**Figure 9.** Proposed process flow sheet for As(V) removal by using Red Mud (Taking into consideration the results of our previous studies,<sup>[6,7]</sup> the section indicated by dashed line may be proposed).

At the end of the serial studies performed in our laboratory, it is demonstrated that both liquid and solid phases of red mud could separately be used in arsenic removal from aqueous solutions, the former is a coagulant and the latter is an adsorbent. As a consequent, following process flow sheet (Fig. 9) may be recommended where all the materials used are each waste- or by-product.



## CONCLUSIONS

The results of the present study demonstrate that the treatment of arsenical wastewaters by coagulation with liquid phase of red mud (LPRM) which is bauxite processing residue is a cost-effective method. Without doing any quantitative assessment, this conclusion is arrived at based on the fact that the basic materials, LPRM, from which  $\text{AlO}_2^-$  is present is cost-free and auxiliary material,  $\text{CO}_2$ , is inexpensive. Compared to the most commonly used coagulants such as alum and ferric salts which are relatively costly and not commonly and readily available, red mud is discarded as a waste product from aluminum industry which could be easily supplied.

From the results of present study the following conclusions can also be drawn.

- (a) When the mixture of LPRM and arsenic containing solution in high alkalinity is neutralized with acid solution accompanied with agitating by air, As(V) co-precipitates with aluminum hydroxide. The efficient As(V) removal is obtained at the final pH range of 5.5–8 for a LPRM/(As(V) solution) ratio of 1/5 (v/v). In this case, more than 90% As(V) is removed from the solution the As(V) concentration of which is  $50 \text{ mg dm}^{-3}$ .
- (b) It was found that the As(V) removal strongly depended on the amount of aluminum precipitated. The Al/As(V) molar ratio is required to be 6 to 8 for efficient As(V) removal from the solutions in different initial As(V) concentrations.
- (c) It was showed that  $\text{CO}_2$  gas can be used instead of acid solution in order to neutralize the mixture of LPRM-As(V) solution. Agitation is also fulfilled when  $\text{CO}_2$  is used as neutralizer. In the  $\text{CO}_2$  neutralization runs, the Al/As(V) molar ratio values are observed to be similar to those of the acid neutralization method. Up to the arsenic concentration of  $20 \text{ mg dm}^{-3}$ , 0.1 volumetric ratio of LPRM/(As(V) solution) is found to be sufficient for efficient As(V) removal.

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