Predicting arsenic behavior in the wastewater of Mouteh Gold Plant: geochemical modeling

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Abstract

Arsenic is one of the heavy metals and nearly all its compounds, especially organic compounds, are toxic. The wide spectrum of diseases caused by this element has led to the evaluation of toxicity of different arsenic species and identification of the major natural and anthropogenic pollution sources of it in nature. Mining activities are among the main sources of anthropogenic pollution of soil and water by arsenic. Using geochemical modeling of different arsenic species in the wastewater of the tailings dam of Mouteh Gold processing plant in Iran, this paper aims to evaluate the effect of pH and temperature on the stability of these components. Modeling was done using MINTEQ software. The results showed that arsenic species at different pH values under study were H_3AsO_3, $H_2AsO_3^-$ and $HAsO_3^{2-}$, and their actual concentration in the plant wastewater were negligible. MINTEQ software introduced H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-} as arsenic V species at different pH values, of which $HAsO_4^{2-}$ and AsO_4^{3-} were the main components of arsenic in plant wastewater. Given the low toxicity of arsenic V species and their easier elimination relative to arsenic III species, in the current conditions, the plant wastewater is in a good status in terms of arsenic pollution. Moreover, temperature changes have little effect on the concentration of various arsenic species in the wastewater; therefore, it is predicted that climatic changes in different seasons have no pollution effect on the wastewater. The results also showed that ytterbium and barium have higher affinity to form precipitates with arsenic and prevent saturation of other species in the solution by removing arsenic.

Keywords: *Arsenic, Wastewater, Geochemical modeling, MINTEQ software.*

1. Introduction

Expansion of modern industry and technology has led to a significant increase in generation and release of pollutants in the environment. Heavy metals, present in the wastewater of many industries, are among the most important pollutants and the most serious risks facing the survival of organisms due to their stability and lack of biodegradation [1].

Minerals processing industry is one of the most productive and effective industries in the release of pollutants due to the large volume of water consumed in this industry. The presence of different heavy metals along with a variety of processed ores generates high volumes of wastewater containing these pollutants. In order to prevent emission of these toxic pollutants by their transfer to water resources and the environment, we should neutralize those using appropriate methods.

Tailings dams are among the focuses of underground water contamination. Availability of minerals, sufficient moisture and oxidative conditions cause faster dissolution of heavy metals in tailings dams, resulting in soil and water pollution. Therefore, these dams are suitable places for wastewater neutralization and precipitation in order to prevent contamination of water resources [2-4].

Arsenic is one of the heavy metals which are usually present in tailings dams of sulfide

processing plants. The major arsenic-containing minerals include arsenopyrite (FeAsS), Realgar (As_4S_4) and Orpiment (As_2S_3), which are abundant in sulfide deposits like gold containing sulfide deposits. It is also found in pyrite, chalcopyrite, galena and sphalerite [5-9].

Typical concentration of arsenic in water is 0.1-80 μg/l [10]. The FAO health limit for arsenic in drinking water had been set at 50 μg/l. This value has been reduced to 5-10 μg/l in the recent reports about arsenic toxicity Exposure since this element causes diseases such as keratosis, skin cancer, testicular and bladder cancer, lung cancer, gangrene, high blood pressure, heart disease and serious damage to kidneys, lungs and liver. The trivalent arsenic can cause nerve cell damage even at concentrations as low as 10 μg/l [5, 6, 11, 12].

The main species of arsenic in aqueous medium are arsenite (H_3AsO_3)and arsenate (H_3AsO_4), containing arsenic III and V, respectively (Nriagu et al., 2007). Pentavalent arsenic can be found in four forms: $H_3AsO_4, H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-}, depending on the pH of the aqueous medium. Similarly, trivalent arsenic is found in H_3AsO_3, $H_2AsO_3^-$ and $HAsO_3^{2-}$ forms. Eh-pH diagram of arsenic species is shown in Figure 1. Ion species of pentavalent arsenic are stable at pH higher than 3, while ion species of trivalent arsenic exist at pH higher than 9 [5, 13].
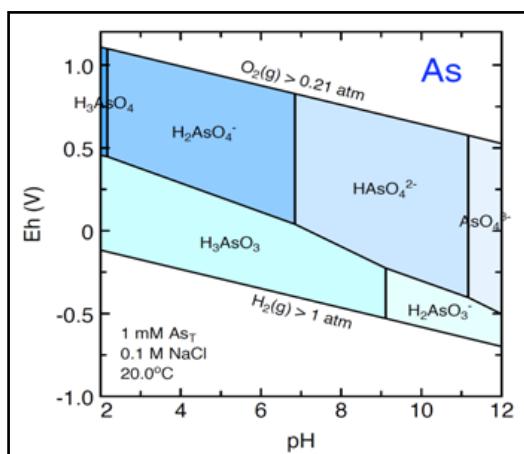

Figure 1. Eh-pH diagram for arsenic species [14].

The trivalent arsenic species are hardly removable because they are neutral in conventional pH values. Pentavalent arsenic species are negative and are, therefore, easier to remove. Trivalent arsenic has thus more mobility in soil and water medium than pentavalent arsenic [5].

Different methods are used to remove arsenic from water. They includeprecipitation, adsorption, ion exchange, membranes, reverse osmosis, biological processes, etc. The refining process of

groundwater by each of these methods demands a lot of time and cost. Therefore, a more prudent approach is to prevent the release of arsenic into the groundwater [10].

The Mouteh gold plant (Figure 2) in Isfahan Province is one of the biggest gold processing projects in Iran. In this plant, gold is extracted from the ore by cyanidation process. Then cyanide complex is absorbed on the activated carbon surface in the presence of oxygen in an alkaline environment. Type of processed ore and operational processes result in the transport of various heavy metals such as cyanide and arsenic to the wastewater, and their accumulation in tailings dam causes long-term negative environmental effects.

Figure 2. Geographic location of Mouteh Gold Plant.

The behavior of arsenic in wastewater depends on a number of parameters such as pH, temperature, etc. Also the capability of forming different complexes of arsenic with other species in wastewater can have a great role in its transfer. Geochemical modeling can be helpful in explanation of neutralization mechanisms of this heavy metal.

In this study, geochemical modeling of various compounds of arsenic in the wastewater of tailings dam in Mouteh gold plant was studied at different pH and temperature conditions to find suitable conditions for neutralization, reduce pollution and prevent the release of this pollutant to the groundwater.

2. Theoretical investigation
2.1. Geochemical formulation

In the chemical model, we have the species (c_1, c_2, c_3, ..., c_{naq}, s_1, s_2, s_3, ..., s_{ns}, p_1, p_2, p_3, ..., p_{np}),

where c_i's are concentrations in aqueous phase; s_n's adsorbed on the solid phase, and p_i's precipitated on the solid phase. The formation of species c_i in the aqueous phase or species s_i and p_i in the solid phase can be described by the mass action law as follows [15].

$$c_i = K_{Ci} \prod_j^{na} X_j^{a_{ij}^a} \quad i = 1, 2, ..., n_a \quad (1)$$

$$s_i = \frac{\rho c_i}{n} = k_{s_i} \prod_j^{n_s} X_j^{a_{ij}^s} \quad i = 1, 2, ..., n_s \quad (2)$$

$$K_{sp_i} \geq \prod_j^{np} X_j^{a_{ij}^p} \quad i = 1, 2, ..n_p \quad (3)$$

where:

K_{ci}= equilibrium formation constant for species c_i
K_{si}=equilibrium formation constant for species s_i
K_{spi}= solubility product for species p_i
X_j= activity of component j
a_{ii}^a= stoichiometry coefficient of component j in species c_i
a_{ii}^s= stoichiometry coefficient of component j in species s_i
a_{ii}^p= stoichiometry coefficient of component j in species p_i
n_a= number of species in the aqueous phase
n_s= number of species adsorbed in the solid phase
n_p= number of species precipitated in the solid phase

In this formulation sorptive sites are treated as one of the many components, (X_i). The mass balance equation requires that the sum of mass for each species in both aqueous and solid phases must be equal to the total amount of mass in the system. Hence, the total concentration of each component is calculated by Eq. 4 or Eq. 5.

$$T_j = C_j + S_j + P_j \quad (4)$$

$$T_j = \sum_i^{n_a} a_{ik}^a c_i + \sum_i^{n_s} a_{im}^S s_i + \sum_i^{n_p} a_{il}^P p_i \quad j = 1, 2, ..n_c \quad (5)$$

where:

Tj = total concentration of component j (M/L³ fluid)
Cj = concentration of component j in the aqueous phase (M/L³ fluid)
Pj = total concentration of component j precipitated on the solid phase (M/L³) or total concentration of component j on the solid phase (M/M solids)
Sj = total concentration of component j adsorbed on the solid phase (M/L³ fluid)
∑= product operator over all components in species j

2.2. Solution method

Geochemical modeling uses the simultaneous solution of the nonlinear mass action expressions and linear mass balance relationships to formulate and solve the multiple-component chemical equilibrium problems. It uses mass-law equations and formation constants for the set of species and material balance equations for each component to define the chemical equilibrium problem. In the model, from an initial guess of the activity of each component (non complex), the provisional concentrations of each species are computed through the mass action expressions written in terms of component activities (Eq. 3). The total mass of each component is then calculated from the concentrations of every species containing that component (Eq. 4). The calculated total mass for each component is then compared with the known input total mass of each component. If the calculated total mass component differs from the input total mass for any component by more than a pre-set tolerance level with the total mass, a new estimate of the component activity is made and the entire procedure is repeated. Having equilibrated the aqueous phase, the model computes the saturation index (SI) for each possible solid with respect to the solution. The solid with the most positive SI is allowed to precipitate. The reverse process occurs if an existing solid is found to be under-saturated with respect to the solution [16].

2.3. MINTEQ software

This software is an equilibrium speciation model that can be used to calculate the equilibrium composition of dilute aqueous solutions in the laboratory or in natural aqueous systems. The model is useful in calculating the equilibrium mass distribution among dissolved species, adsorbed species, and multiple solid phases under a variety of conditions including a gas phase with constant partial pressures. A comprehensive data base is included that is adequate for solving a broad range of problems without any additional user-supplied equilibrium constants [15,16].

3. Tailings dam characteristic of Mouteh gold plant

The wastewater sample which was taken from the underflow sludge of the thickener in the Mouteh gold processing plant was precipitated and filtrated. This sample was analyzed by ICP method. Table 1 shows the results of the analysis of this solution.

As shown in Table 1, the arsenic species in the sample is AsO_4^{3-} (Pentavalent arsenic), which is proportional to the high pH of the tailings dam wastewater (Figure 1). The high concentration of

this species in wastewater (427 mg/L) that is much more than the permitted limit (0.01 mg/L [17]) provides enough motivation for this research.

4. Results and discussion

For geochemical modeling of arsenic in the effluent, the data in Table 1 were fed into the software as initial values. Then the Extended Debye Huckel equation was used for calculation of the reactants concentrations and correction of the activities (i.e. conversions between molality and activity). This equation provides acceptable activity coefficients for ionic strengths. In electrochemical calculations, the H_3AsO_3/AsO_4 pair was considered. The most important results of this modeling were presented and analyzed to examine the behavior of arsenic species at different pH values and temperatures.

4.1. The minerals formed

A list of the minerals formed in the MINTEQ software model is given in Table 2. As can be seen, among the minerals formed in the software model, $BaHAsO_4:H_2O$ (s), $Co_3(AsO_4)_2$ (s), $Zn_3(AsO_4)_2:2.5H_2O$ (s) and $YbAsO_4$ (s) are arsenic containing minerals. The very low concentration of ytterbium in the input data (7.4 mg/L) indicates its high affinity for arsenic.

Table 1. The concentration of ions in the wastewater of Mouteh tailings dam

Ions	Concentration (mg/L)	Ions	Concentration (mg/L)	Ions	Concentration (mg/L)
H^+	0	Cs^+	3.99	Br^-	38
CO^{2+}	61.99	Fe^{2+}	566	Cr^{2+}	32
CN^-	270	Na^+	2.27	Zn^{2+}	196
AsO_4^{3-}	427	Rb^+	160	Yb^{3+}	7.4
Ba^{2+}	1800	$Sb(OH)_3$	1.8	-	-

Table 2. Minerals formed in MINTEQ software model

No	Minerals	No	Minerals
1	$Ba(OH)_2:8H_2O$ (s)	15	Senarmontite
2	$BaHAsO_4:H_2O$ (s)	16	Valentinite
3	$Co(OH)_2$ (am)	17	$Yb(OH)_3$ (s)
4	$Co(OH)_2$	18	$YbAsO_4$ (s)
5	$Co_3(AsO_4)_2$ (s)	19	Zincite
6	CoO(s)	20	$Zn(OH)_2$ (am)
7	$Cr(OH)_2$ (s)	21	$Zn(OH)_2$ (beta)
8	$Fe(OH)_2$ (am)	22	$Zn(OH)_2$ (delta)
9	$Fe(OH)_2$	23	$Zn(OH)_2$ (epsilon)
10	NaCN (cubic)	24	$Zn(OH)_2$ (gamma)
11	Sb_2O_3 (s)	25	$Zn_2Fe(CN)_6$ (s)
12	Sb_4O_6 (cubic)	26	$Zn_3AsO4_2:2.5H_2O$ (s)
13	Sb_4O_6 (rhombic)	27	$ZnBr_2:2H_2O$ (s)
14	$SbBr_3$ (s)	28	$Zn(OH)_2$ (delta)

4.2. Estimation of equilibrium pH at different temperatures

Since the minimum and maximum temperatures in the Mouteh gold mine are 10 and 40°C, respectively, the values 10, 25 and 40°C were used as the temperature levels in the software. Table 3 shows the equilibrium pH values in the selected temperatures calculated by the software using mass balance calculations.

4.3. Modeling soluble arsenic III species at different pH values

The modeling results introduced H_3AsO_3, $H_2AsO_3^-$ and $HAsO_3^{2-}$ as the species of arsenic III detected in the wastewater. Figures 3-5 show the concentration of these species at different pH values and selected temperatures. As shown clearly, the typical trivalent arsenic component, especially at pH values less than 7, is H_3AsO_3, which is also confirmed by the Eh-pH diagram shown in Figure 1. However, considering the actual conditions of the wastewater in the plant (pH 11-12), the concentration of all arsenic III species is negligible, which causes no environmental problem.

Table 3. Equilibrium pH values in selected temperatures

Temperatures	Equilibrium pH
10	11.87
25	11.53
40	11.19

4.4. Modeling the soluble arsenic V species at different pH values

The modeling results indicated H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-} as pentavalent arsenic species. Figures 6-8 show the concentration of these species at different pH values and selected temperatures.
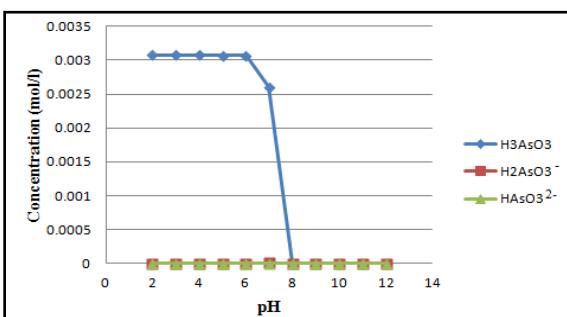

Figure 3. Changes in the concentration of arsenic III species in three different pH values and 10°C temperature.

Figure 4. Changes in the concentration of arsenic III species in three different pH values and 25°C temperature.

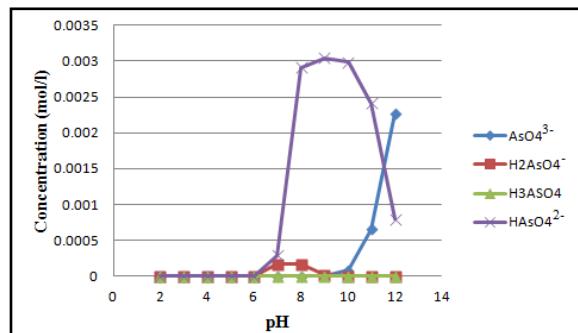**Figure 5. Changes in the concentration of arsenic III species in three different pH values and 40°C temperature.**

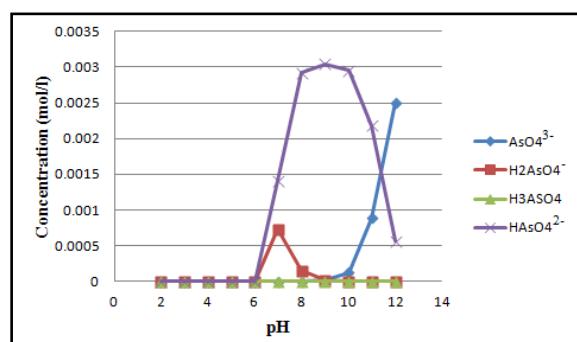**Figure 6. Changes in the concentration of arsenic V species in three different pH values and 10°C temperature.**

Figure 7. Changes in the concentration of arsenic V species in three different pH values and 25°C temperature.
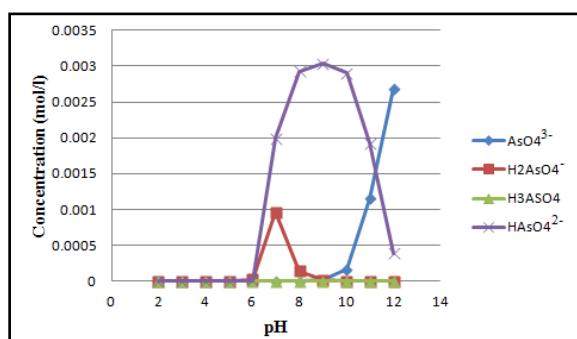

Figure 8. Changes in the concentration of arsenic V species in three different pH values and 40°C temperature.

These diagrams show that H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-} are the main species of arsenic V at studied pH values, and that H_3AsO_4 concentration is negligible. Also regarding the wastewater equilibrium pH (11-12), the concentration of $H_2AsO_4^-$ is very low. So, the major arsenic V species are $HAsO_4^{2-}$ and AsO_4^{3-} that should be highlighted in the monitoring of arsenic in the plant tailings dam. By increasing of pH values above 10 the concentration of $HAsO_4^{2-}$ increases while the AsO_4^{3-} concentration decreases.

4.5. Comparison of the modeling results for soluble species of arsenic III and V

Comparative analysis of the diagrams for species ofarsenic III and V shows that an increase in the pH value decreases the arsenic III species concentration and increases the concentration of arsenic V species. Unlike the arsenic III species, negative species dominating species of arsenic V. Non-ionic species have less mobility and are more removable compared to ionic species [5]. Therefore, arsenic causes more environmental problems in lower pH values than higher pH levels.

It is worth noting that, in addition to soluble species of arsenic III and V, a number of arsenic species were also observed in the results of modeling which have great tendency to precipitate, resulting in partial arsenic removal from the solution.

These compounds include arsenolite, $BaHAsO_4.H_2O$ (s), As_2O_5 (s), $YbAsO_4$ (s), $Co_3(AsO_4)_2$ (s) and $Zn_3AsO_4:2.5H_2O$ (s).

4.6. The effect of temperature on soluble species of arsenic III and V

In this section, the effect of temperature on arsenic III and V species in wastewater has been assessed. H_3AsO_3, $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-} have been, respectively, considered as typical arsenic III and V species because they have the highest concentrations in the studied pH values. Figures 9-12 show the changes in the concentration of these species at different pH values and selected temperatures.

As shown in Figures 9-12, temperature changes do not cause major changes in the species of arsenic III and V, but in general, increasing of temperature reduces the concentration of typical species of arsenic III and increases the concentration of some typical species of arsenic V.
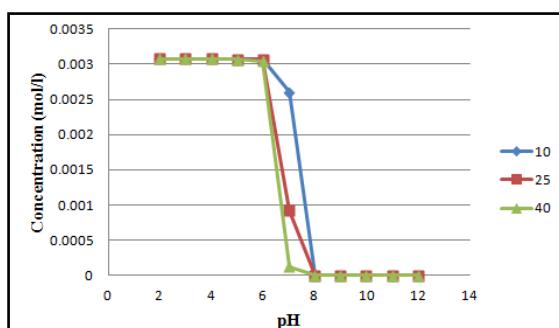

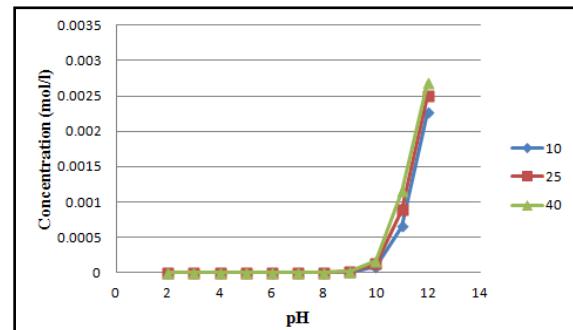**Figure 9. Changes in the concentration of H_3AsO^3 species with increasing temperature at different pH values.**

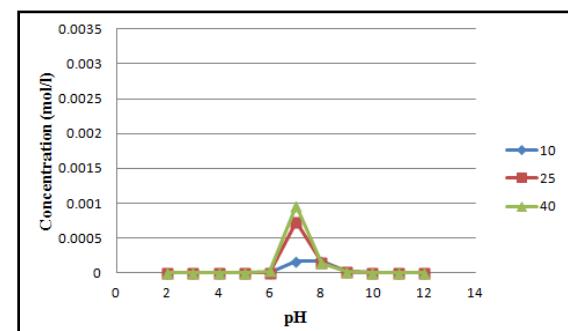**Figure 10. Changes in the concentration of AsO_4^{3-} species with increasing temperature at different pH values.**

Figure 11. Changes in the concentration of H_2AsO^{4-} species with increasing temperature at different pH values.
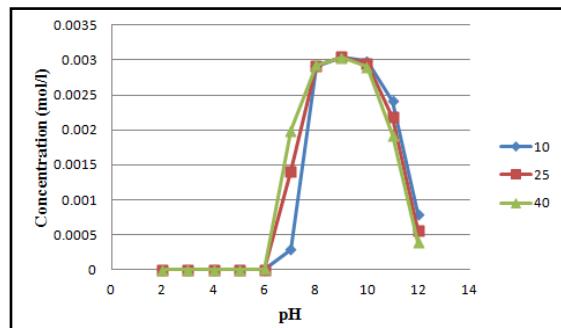

Figure 12. Changes in the concentration of $HAsO_4^{2-}$ species with increasing temperature at different pH values.

4.7. Evaluation of the Saturation Index (SI) of the solid species of arsenic at different pH values

Saturation index is an appropriate criterion for comparing tendency of different metals to precipitate with arsenic, and is defined as bellow:

$$SI = \log \frac{IAP}{K_s} \qquad (9)$$

where, IAP and K_s are ion activity and solubility constant, respectively. A positive SI value suggests that precipitation occurs and the solution is supersaturated relative to the solid. For SI value equal to zero, the solution is in an apparent

balance relative to the solid and when SI is negative, the solution is undersaturated relative to the solid.

When precipitation possibility is considered for super saturated solid species, the species precipitate in the solution and saturation index reaches zero. Unless the solution is undersaturated relative to the solid, its saturation index will be lower than zero [15,16].

Figures 13-14 were plotted for the solid species of arsenic in the two states including with and without precipitation.
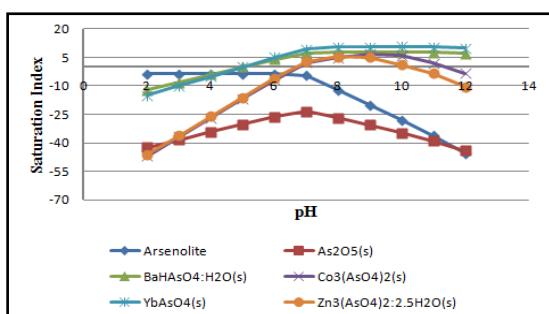

Figure 13. Changes in the saturation index of the solid species in different pH values (excluding precipitation).
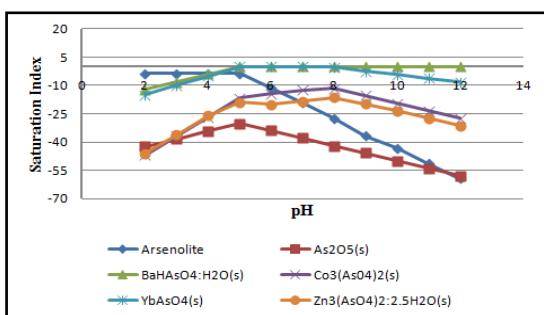

Figure 14. Changes in the saturation index of the solid species in different pH values (including precipitation).

As can be seen in Figure 13, without precipitation, there is the possibility of super- saturation for the four species of $YbAsO_4$ (s), $BaHAsO_4$: H_2O(s), $Co_3(AsO_4)_2$ (s) and $Zn_3AsO_4:2.5H_2O$ (s), but when precipitation occurs in Figure 14, only the two solid species of $YbAsO_4$ (s) and $BaHAsO_4:H_2O$ (s) do precipitate, causing $Co_3(AsO_4)_2$ (s) and $Zn_3(AsO_4)_2:2.5H_2O$ (s) species to become undersaturated. These solid species precipitate after reaching the saturation index of zero and remove arsenic from the solution (Figure 15); this indicates that the elements barium and ytterbium have a higher ability to form solid compounds with arsenic than cobalt and zinc. In fact, by formation of solid arsenic compounds with barium and ytterbium and their elimination from the solution, the possibility of supersaturated state for cobalt and zinc with arsenic is removed. Another

important point is that, in spite of much lower concentration of ytterbium relative to barium, zinc and cobalt (7.4 against 1800, 196 and 62 mg/L), the former shows higher ability to form solid compounds with arsenic.

Figures 16-18 show changes in typical species concentration of arsenic III (H_3AsO_3) and V ($HAsO_4^{2-}$ and AsO_4^{3-}) at different pH values and in the two states of with and without precipitation.

As shown in Figure 15, in precipitation state, the H_3AsO_3 species faces a sharp drop in concentration in the pH range of 6-8. According to Figure 13, in this range of pH, arsenic V precipitates including $YbAsO_4$ (s) and $BaHAsO_4:H_2O$ (s) are formed, removing a major portion of total arsenic in solution (Figure 14).
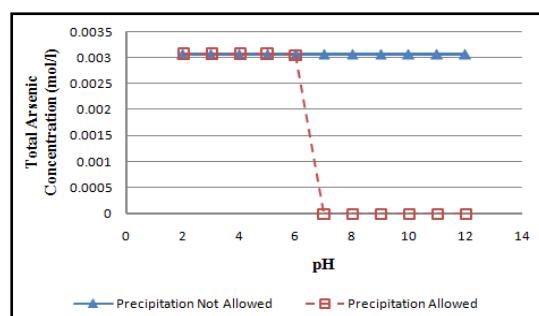

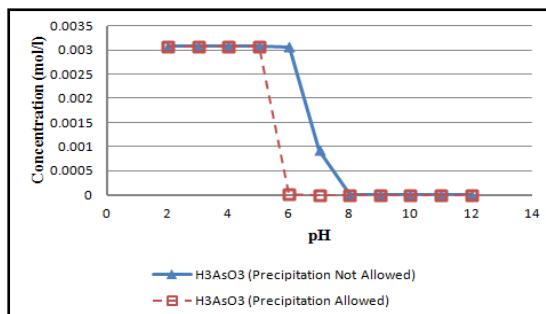**Figure 15. Changes in the total arsenic concentration at different pH values with and without precipitation.**
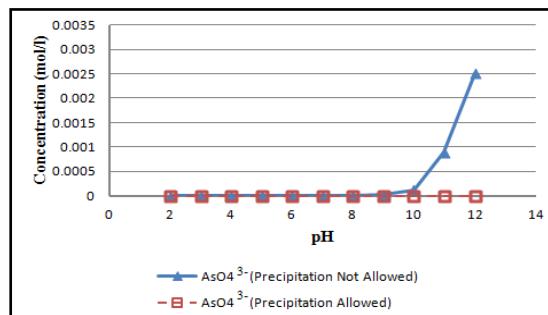

Figure 16. Changes in the concentration of H_3AsO_3 species at different pH values (with and without precipitation).

Figure 17. Changes in the concentration of AsO_4^{3-} species at different pH values (with and without precipitation).
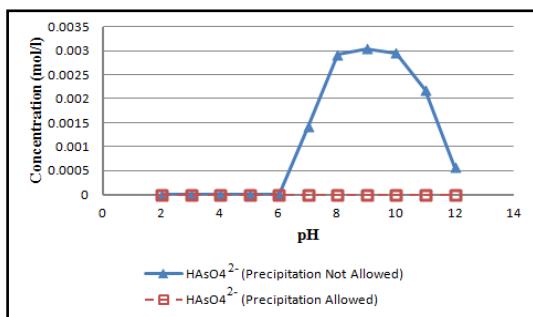
Figure 18. Changes in the concentration of $HAsO_4^{2-}$ species at different pH values (with and without precipitation).

Consequently, decrease of arsenic III species concentration in solution occurs by their oxidation to arsenic V species, in order to satisfy chemical equilibrium between these species in solution.

In Figure 16, a sharp drop in AsO_4^{3-} concentration is visible at pH values above 9. According to the figures in previous sections, AsO_4^{3-} is one of the two species of arsenic in the mentioned pH values, and also based on Figure 13, the only precipitate that can be formed in this pH range is $BaHAsO_4$: H_2O (s). Therefore, it is obvious that the decrease in the AsO_4^{3-} concentration is related to the formation of the mentioned precipitate.

The great decrease of $HAsO_4^{2-}$ concentration in the pH values higher than 6 (Figure 17) is due to formation of $BaHAsO_4:H_2O$ (s) and $YbAsO_4$ (s) precipitations and consequently, significant reduction of total arsenic in solution (Figure 14).

5. Conclusions

The results of geochemical modeling of arsenic in the tailings dam of the Mouteh gold processing plant indicate that:

- MINTEQ software is a proper tool to introduce various species of arsenic in wastewater to identify effective factors in the removal of this contaminant.
- Arsenic III species in the studied pH values are H_3AsO_3, $H_2AsO_3^-$ and $HAsO_3^{2-}$, and their concentration is negligible in the actual conditions of the plant wastewater.
- MINTEQ software introduces H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-} as arsenic V species at different pH values, of which $HAsO_4^{2-}$ and AsO_4^{3-} are major species in the actual conditions of the plant wastewater.
- Generally, the plant wastewater is currently in good condition in terms of arsenic contamination, because present arsenic species in the wastewater are arsenic V species which can be easily removed and are less toxic.
- Climatic temperature changes in different seasons have no considerable effect on the concentration of arsenic species in wastewater.
- Among the elements presented in the input data, ytterbium and barium have higher affinity to form precipitates with arsenic which prevent saturation of other species in the solution by removing arsenic.

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