

Detoxification of Arsenic-Containing Wastewater Using Bauxite and Clay*

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Abstract

Arsenic is a very toxic substance, and one of the priority pollutants in waste discharges. Mining companies that treat ores containing arsenic-bearing minerals end up releasing arsenic into their tailings impoundment. Arsenic can cause harm should it find its way into soils, streams, and groundwater. In this study, adsorbents at ratios of 80%, 60% and 50% bauxite to clay were used to remove arsenic from solution at initial arsenic concentrations of 20 ppm, 10 ppm, and 5 ppm. The effect of adsorbent dosage and initial arsenic concentration on arsenic removal efficiency were studied. The results indicated higher removal of arsenic from the 20-ppm solution for all the bauxite/clay mixtures, as compared to the lower arsenic concentration solutions. Whereas arsenic removal from the 20-ppm solution was favoured by 80% bauxite in the bauxite/clay mixture, the 5-ppm arsenic concentration was sorbed more by the bauxite/clay mixture when the clay content was increased to 50%. The higher amounts of As sorbed with increasing percentage of bauxite can be attributed to the presence of gibbsite and goethite. The adsorption kinetics models were studied, and the results suggest that the reaction was chemisorption as the pseudo-second-order appeared to fit the data more than the pseudo-first-order. Though the general trends obtained appears inconclusive, the results from this work establishes the feasibility of incorporating bauxite and clay into the design of a tailings impoundment facility for arsenic removal.

Keywords: Arsenic; Bauxite; Clay; Adsorption; Kinetics

1 Introduction

Management of hazardous wastes is of a major public concern, and arsenic is one of the priority pollutants in waste discharges. Arsenic is introduced into aqueous systems through geochemical reactions, industrial waste discharges and agricultural use of arsenical pesticides, among others. Naturally, arsenic-bearing minerals in ore deposits are usually stable in their undisturbed state. Exposure to oxidising environments from mining and mineral processing often leaves arsenic in a less stable state, which creates opportunities for off-site transport, usually by water (Loukidou *et al.*, 2003; Alshaebi *et al.*, 2009).

Arsenic present in water is mainly in the forms of arsenate (As(V)) and arsenite (As(III)), which are (AsO_4^{3-}) and (AsO_3) respectively. In a typical environment at a pH range of 2 - 12, the dominant As(V) species are negatively charged (H_2AsO_4^- and HAsO_4^{2-}), while the dominant As(III) species is neutral (H_3AsO_3) and predominates under anaerobic conditions within a pH range of 2 to 9. The negatively charged As(V) species are more likely to be adsorbed and are generally more easily removed than As(III) in treatment systems (Kumar *et al.*, 2004; Parga *et al.*, 2005; Roghani *et al.*, 2016; Uppal *et al.*, 2016; Qin *et al.*, 2017; Ozola *et al.*, 2019). At typical pH values (pH 5-8) in natural waters, arsenate exists as an anion while arsenite remains a

fully protonated molecule (Rubidge, 2007). The toxicity of arsenic in aquatic environments depends on its speciation, solubility, and mobility, as well as its stability (Karmacharya *et al.*, 2016; Foroutan *et al.*, 2017). Health effects of arsenic are normally characterised according to the dose of the particular arsenic compound. The permitted concentration by WHO and EPA for drinking water is 10 $\mu\text{g/L}$ (Anon, 2020; Goswami *et al.*, 2012; Anon, 2021). Arsenic toxicity causes skin lesions and bladder cancers, and damages mucous membranes, digestive, respiratory, circulatory and nervous systems (Mamisahebei *et al.*, 2007; Rubidge, 2007).

Arsenic may be adsorbed by numerous materials including phyllosilicates, silica, hydrous oxides of iron and aluminum, treated activated carbon, treated zeolites, sea sand, magnesium hydroxide, and manganese dioxide (Rubidge, 2007). To remove arsenic from aqueous solutions and wastewaters, numerous techniques including physical-chemical methods such as flocculation, filtration, oxidation-reduction, electrochemical methods, ion exchange, reverse osmosis, electrodialysis, iron ore sludge, bauxite, clay, and adsorption have been employed (Barakat *et al.*, 2013; Adeleye *et al.*, 2016; Foroutan *et al.*, 2017; Silva-Yumi *et al.*, 2018; Bien and Ha 2019, Ndur *et al.*, 2019).

Currently, there is great interest in using bauxite, which is relatively cheaper, to adsorb arsenic from

such effluents and other wastewaters. Bauxite is a naturally-occurring, heterogeneous material composed primarily of one or more aluminium hydroxide minerals as the main constituent, with various mixtures of silica, iron oxide, titania, aluminosilicate, and other impurities in minor or trace amounts. The principal aluminium hydroxide minerals found in varying proportions with bauxites are gibbsite $\{Al_2O_3 \cdot 3H_2O$ (Al_2O_3 -65.4%; Al-34.6%)}, the polymorphs boehmite and diaspore $\{Al_2O_3 \cdot H_2O$ (Al_2O_3 -85%; Al-45%)}, and colloidal alumina hydrogel $\{Al_2O_3 \cdot 2H_2O$ (Al_2O_3 -73.9 %; Al-39.1%)}

 (Alshaebi *et al.*, 2009; Ostojić *et al.*, 2014; Ozola *et al.*, 2019).

The use of clay as adsorbent has been investigated by many researchers due to it being relatively cheaper than other adsorbents, its high abundance, and its numerous properties including large specific area, high capacity to exchange cation and anion, and its tendency to adsorb water and exhibit plasticity when mixed with water in various proportions (Nayak and Singh, 2007; Benthahar *et al.*, 2016). Geologic clay deposits are mostly composed of phyllosilicate minerals with a variable amount of water trapped in the mineral structure which is typically formed over long periods with traces of metal oxides and organic matter. Clay surfaces are characterised by a permanent negative charge due to isomorphic substitution, which exhibits low adsorption capacity of some anionic elements (Ren *et al.*, 2014).

Interest in gold ores has led to discoveries where some ores and arsenic are strongly correlated in many deposits, which makes mining industries the major source of arsenic waste. This is especially so with mining companies that treat refractory ores containing arsenopyrite, as arsenic end up being a by-product in the tailings dam. These companies produce arsenic solutions with high concentrations which must be reduced to meet the environmental requirements (Alshaebi *et al.*, 2010). Thus, such companies employ a neutralisation process using limestone and lime to reduce the arsenic levels before discharge into the tailings impoundment (Alshaebi *et al.*, 2010).

This study aimed at assessing the possibility of incorporating bauxite and clay into the design of tailings impoundment to synergistically adsorb arsenic in wastewater to a safe concentration before it is released into the environment. This will help reduce possible contamination of ground and surface waters in the event of seepage, and thus respond to the Sustainable Development Goals 6, 14 and 15.

2 Resources and Methods Used

2.1 Materials Used

Bauxite and clay were used in this study as materials for the removal of arsenic from wastewater. The clay was obtained from the University of Mines and Technology (UMaT) construction site while bauxite was obtained from Awaso. Mineral components were determined using a Philips Xpert diffractometer.

Arsenic trioxide used in the preparation of arsenic solution were obtained from the UMaT Minerals Engineering Laboratory. The plastic container and Polyvinyl Chloride (PVC) pipe were purchased from Tarkwa market and used in the construction of the miniature tailings dam.

2.2 Experimental Set-up

Three holes each of the same diameter (2.5 cm) were made through the plastic container on one side. Three adapters were inserted into the holes, and pieces of latex foam, fixed in them to prevent the clay-bauxite mixture from reporting in the arsenic solution. The detailed construction is shown in Fig.1.

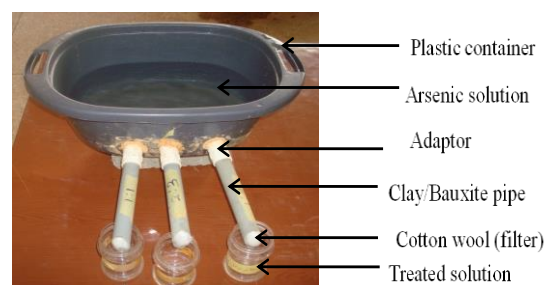


Fig. 1 Experimental Set-up

A PVC pipe of diameter 2 cm was cut into three parts (15 cm) each and inserted in the adapters. The PVC pipes were filled with bauxite and clay of varying ratios, and cotton wool was used to plug the ends as shown in Fig 1 to prevent the clay-bauxite mixture from contaminating the treated solution. A container was placed under each of the three pipes to collect the solution that passed through them.

Five litres of arsenic-containing solutions of concentrations, 20 ppm, 10 ppm, and 5 ppm were prepared. To obtain the above concentrations, 140 mg, 70 mg, and 33 mg of arsenic trioxide (As_2O_3) were weighed respectively into three different containers, and 255 ml of concentrated sulphuric acid (H_2SO_4) was added to make each of the arsenic solutions. The mixture of sulphuric acid and arsenic trioxide was placed on a hot plate to speed up the dissolution, after which the solution was allowed to

cool. The solution lost as vapour was replenished with distilled water.

The bauxite was screened to 1.4-mm particle size for all the bauxite ratios since it had been established by previous studies that adsorption is optimum at particle size less than 2 mm (Essilfie, 2010). The clay was also milled, dried, and agglomerated particle size of 1.18 mm were utilised. The clay to bauxite ratio was varied and the ratios used were 2:3, 1:4, and 1:1. The arsenic solution was poured into the container and allowed to pass through the clay-bauxite mixture-containing pipes and collected into three different containers. Samples of solutions from each container were taken at time intervals of 20 min, 40 min, 60 min, and 120 min.

2.3 Analysis of Results

The kinetic data of As adsorption were suited to model equations (pseudo-first-order and pseudo-second-order) by linear regression using the least-squares method. The correlation coefficients (R^2) obtained from the regression analyses were used to evaluate the applicability of the model equations.

3 Results and Discussion

The removal of arsenic from solution was investigated using mixtures of clay and bauxite as adsorbents. The mineralogical composition of the bauxite and clay utilised are presented in Figs. 2 and 3. The major minerals in the clay were silica (SiO_2) and potassium sodium oxide at 70.4% and 26% respectively. Zinc iron oxide and aluminosilicates were also present in minute quantities. The bauxite was predominantly composed of gibbsite ($\text{Al}(\text{OH})_3$) with 94.5% while goethite (FeOOH) and silica were present at 4.5% and 1% respectively. Iron oxides and gibbsite are known to sorb arsenic (Ndur *et al.*, 2019).

The effect of the initial concentration of the arsenic solution and adsorbent dosage on sorption of arsenic are presented in Sections 3.1, 3.2 and to 3.3. The computations of the percentage of arsenic sorbed were done using Equation 1, where C_o is the initial concentration of the arsenic solution and C_t is the concentration of arsenic in the treated solution at any time t .

$$\% \text{ Adsorbed} = \frac{C_o - C_t}{C_o} \times 100\% \quad (1)$$

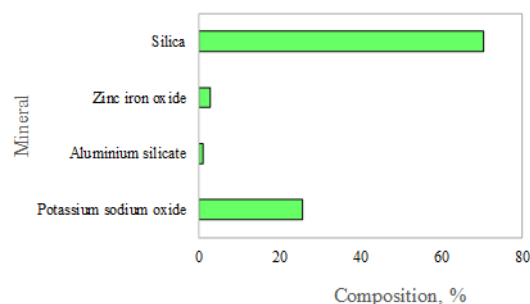


Fig. 2 Mineralogical Composition of Clay Utilised

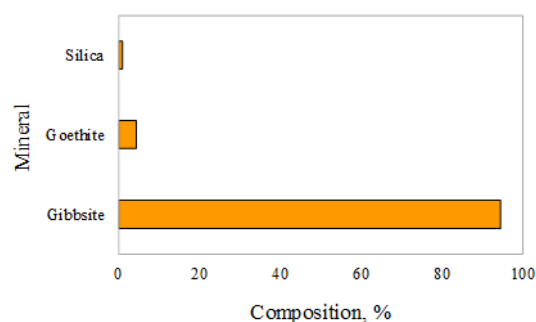


Fig. 3 Mineralogical Composition of Bauxite Utilised

3.1 Effect of Initial Concentration of Arsenic Solution on Sorption by 80% Bauxite /Clay Mixture

Fig. 4 represents the effect of initial arsenic concentration at 20 ppm, 10 ppm, and 5 ppm on sorption using 80% bauxite by clay sorbent within 120 minutes. It was observed that the removal efficiency was highest at 87% for the 20-ppm arsenic at 40 minutes, though the percentage dropped to 82% at 60 minutes, and finally to 78% at 120 minutes. However, for the 10-ppm solution, the percentage of arsenic removed was 29% at 40 minutes; this dropped to 14% at 60 minutes, and increased to 66% after 120 minutes. For the 5 ppm, adsorption was high at 31% after 40 minutes, dropping to 19% after 60 minutes, and finally to 10% at 120 minutes. The 20-ppm had the highest sorption capacity due to the amount of adsorbate present in the solution. The 10 and 5 ppm were lower since the amount of adsorbate decreased and thereby decreasing the concentration gradient for the adsorption to take place, which confirms that removal of arsenic is better in environment where bauxite concentrations are higher (Ndur *et al.*, 2019).

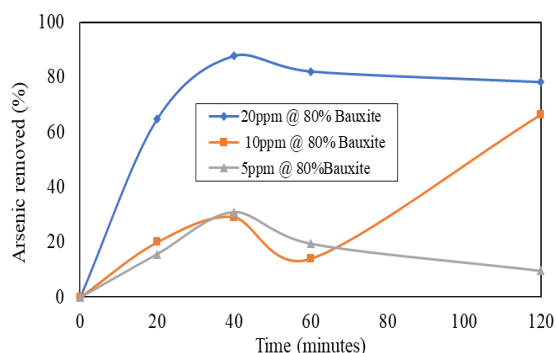


Fig. 4 Arsenic Removal from Solution Using Bauxite-Clay Mixture Containing 80% Bauxite

3.2 Effect of Initial Concentration of Arsenic Solution on Sorption by 60% Bauxite /Clay Mixture

Fig. 5 shows a rise in the adsorption of 20-ppm As solution between time 0 and 60 minutes where 68% of arsenic was sorbed, but gradually decreased to 56% which also confirms the behaviour as observed in Fig. 4.

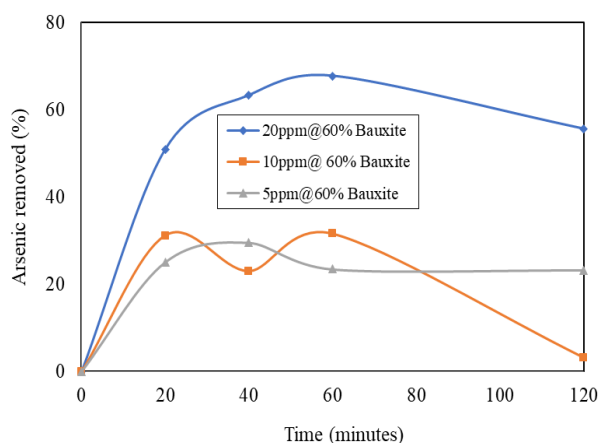


Fig. 5 Arsenic Removal from Solution Using Bauxite-Clay Mixture Containing 60% Bauxite

For the 10 ppm, the percentage of arsenic removed increased to 31% after 20 minutes and then dropped to 23% after 40 minutes, increasing again to 32% and finally down to 3% at 120 minutes. In the case of the 5 ppm, however, 30% was removed after 40 minutes, beyond which it dropped and remained at 23% until 120 minutes. The observations here corroborate those in Fig. 2 that the concentration gradients were low for adsorption from the 5 ppm and 10 ppm.

3.3 Effect of Initial Concentration of Arsenic Solution on Sorption by 50% Bauxite /Clay Mixture

The effect of initial arsenic concentration at 20 ppm, 10 ppm, and 5 ppm on sorption using bauxite-clay mixture containing 50% bauxite is depicted in Fig. 6, where there was a rise in the sorption abilities of bauxite and clay in the initial minutes. This figure shows that the percentage of clay added to bauxite had influence on the active sites of the mixture, which contributed to the high change in sorption ability in the initial minutes of the process. The percentage of arsenic removed from the 20-ppm increased to 73% in 20 minutes and then dropped to 45% in 40 minutes, increasing again to 79% after 60 minutes and finally dropping to 55% in 120 minutes.

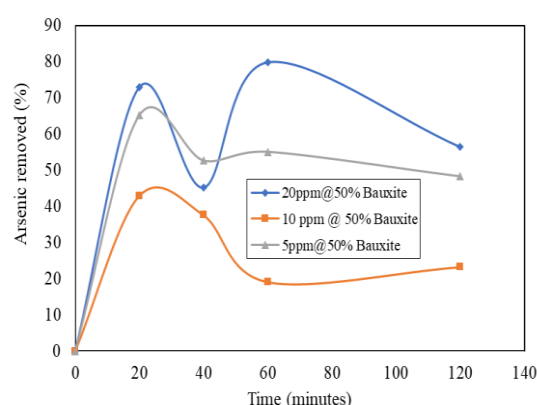


Fig. 6 Effect Arsenic Removal from Solution Using Bauxite-Clay Mixture Containing 50% Bauxite

For the 10 ppm, the percentage of arsenic removed increased to 43% and then decreased to 23% in 120 minutes. For the 5 ppm, the percentage of arsenic removed increased to 65% in 20 minutes, after which it dropped to 48% by the 120-minutes. In general, for most of the systems, arsenic removal followed a sinusoidal trend, due to adsorption and desorption of arsenic ions from and into the solution.

3.4 Combined Effect of Initial Concentration and Dosage of Sorbent

The summary of the combined results is shown in Fig. 7, and the bars represent the highest sorption obtained in each case. For the 80% bauxite, the highest percentage removal of arsenic was 87% for the 20-ppm, followed by 66% for the 10-ppm, and the least percentage of 31% was removed when the concentration was 5 ppm. This can be attributed to the fact that more adsorption sites were available, coupled with a high concentration of the adsorbate in the solution for the 20 ppm and 10 ppm. The higher amounts of As sorbed with increasing percentage of bauxite can be attributed to the

presence of gibbsite and goethite. Inferences from Figs. 4, 5 and 6 also give indications of the contact times that would be required for maximum sorption for each of the set-ups, if this process is to be applied. For example, for 20 ppm arsenic solution and 80% bauxite, 40 min gives the maximum sorption.

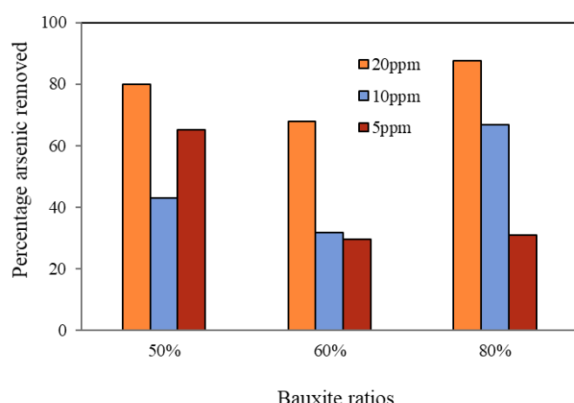


Fig. 7 Combined Effect of Initial Concentration and Dosage of Sorbent

For the 50% bauxite, the percentage of arsenic removed was 79% for the 20 ppm, followed by 65% for the 5 ppm and finally 43% for the 10 ppm. The 50% bauxite recorded the second-highest adsorption percentages for the 5 and 20 ppm except for the 10 ppm which was higher in 80% bauxite. This could be due to the increase in clay content of the mixture which was favoured by the increase in residence time. For the 60% bauxite, the highest percentage of arsenic removed was 68% for 20 ppm, followed by 31% for 10 ppm, and the least was recorded at 29% at 5 ppm. The 60% bauxite recorded the least percentage of arsenic removed for all the concentrations. It can be inferred from the figures that for higher concentrations of the arsenic solution, a higher percentage of bauxite in the bauxite/clay mixture is more favorable, whereas, for the lower concentrations, the amount of clay in the mixture has to be increased. This could be attributed to an increase in the adsorptive surface area, and greater accessibility of surface binding sites to the composite adsorbent clay (Joshi *et al.*, 2019).

3.5 Kinetics Model of Arsenic Adsorption by 80% Bauxite/Clay

The sorption kinetics of As onto the 80% bauxite with 20% clay is shown in Fig 4 and Fig 7. The kinetics of arsenic adsorption onto bauxite and clay was evaluated using two kinetic models to mathematically describe the adsorption constant, though the 50% Bauxite/clay gave a good sorption capacity for low concentration of As. The linearised forms of pseudo-first-order (PFO) and pseudo-second-order (PSO) are expressed in Equations 2

and 3 (Blanchard *et al.*, 1984; Santhy and Selvapathy, 2006; Bulut and Ozacar, 2008):

$$\log \log (q_e - q_t) = \log \log (q_e) - \left(\frac{k_1}{2.303} \right) t \quad (2)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \left(\frac{1}{q_e} \right) t \quad (3)$$

In the equations, q_t and q_e are solid-phase ion concentrations at any time (t) and equilibrium (mg/g), respectively; $K_1(\text{min}^{-1})$ and $K_2(\text{mgg}^{-1}\text{min}^{-1})$ are the rate constants of pseudo-first-order and pseudo-second-order models respectively.

Fig. 8 shows the values of the R^2 for pseudo-first-order sorption kinetics with only 10 ppm having the highest value of 0.6172 with that of 5 and 20 ppm below 0.5 whereas Fig. 9 shows a higher R^2 values for 5 ppm and 20 ppm with 10 ppm having the least R^2 . The results show a direct opposition of sorption kinetics between the first and second kinetics models.

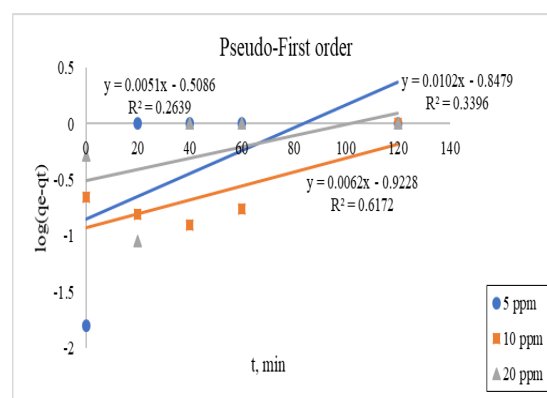


Fig. 8 Pseudo-First-Order Sorption Kinetics Model of As Adsorption onto 80% Bauxite/Clay

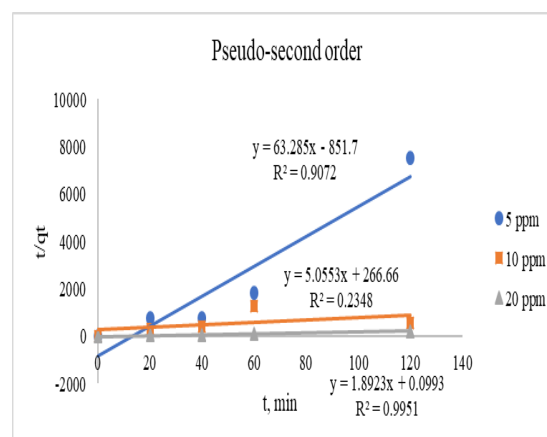


Fig. 9 Pseudo-Second-Order Sorption Kinetics Model of As Adsorption onto 80% Bauxite/Clay

As seen from Tables 1 and 2, the pseudo first-order rate constant k_1 increased with the decrease in the arsenic concentrations (Abechi *et al.*, 2011) whereas the calculated q_e increased with increase in concentration for both the studied adsorption processes. It was also evident that there was a considerable difference between calculated q_e values and the experimental q_e . The values of $K_2(\text{mgg}^{-1}\text{min}^{-1})$ were found to be slightly higher than the $K_1(\text{min}^{-1})$.

Table 1 Pseudo-First-Order Kinetic Model Parameters and Correlation Coefficients for As Sorption

C_0	$q_e(\text{Exp})$	$q_e(\text{Cal})$	$K_1(\text{min}^{-1})$	R^2
5	0.01594	0.1420	0.0235	0.3396
10	0.22091	0.1196	0.0143	0.6172
20	0.5212	0.3100	0.0117	0.2639

Table 2 Pseudo-Second-Order Kinetic Model Parameters and Correlation Coefficients for As Sorption

C_0	$q_e(\text{Exp})$	$q_e(\text{Cal})$	$K_2(\text{mgg}^{-1}\text{min}^{-1})$	R^2
5	0.0161	4.7023	0.9072	0.9072
10	0.1978	0.0958	0.2348	0.2348
20	0.5285	3.5984	0.9951	0.9951

The regression analyses showed that the pseudo-second-order equation appeared to fit the data better than the pseudo-first-order model even though the R^2 (~ 1) for 10 ppm As for both models were far from 1. The better fit for the pseudo-second-order model suggests that the adsorption process might be chemisorption (Jovanović *et al.*, 2011; Nur *et al.*, 2014; Bien *et al.*, 2019).

4 Conclusions

This paper assessed the use of different mixtures of bauxite and clay for the removal of arsenic from solutions of different arsenic concentrations. The results indicated higher removal of arsenic from the 20-ppm solution for all the bauxite/clay mixtures, as compared to the 10-ppm and 5-ppm solutions. Whereas increase in bauxite content to 80% favoured the 20-ppm arsenic concentration, a decrease in bauxite concentration to 50% favoured the 5-ppm arsenic concentration. The kinetic model study suggests that sorption of As onto bauxite/clay is a pseudo-second-order reaction, hence sorption is mainly by chemisorption. It is evident from the study that incorporating bauxite and clay into the design of a tailings impoundment facility is feasible. However, further research is required to validate some of the observations made in this study.

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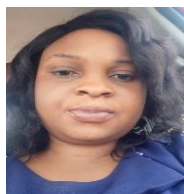
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