



Comparative Study of Adsorption Capacity of Two Mixed Materials for Arsenic Remediation

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Abstract

Arsenic pollution is one of issues for drinkable water supply in rural areas of Burkina Faso. The objective of this study was to look for a cheap technology for a better treatment of enriched arsenic water up to the admissible value (10 µg/L) in drinking water. To fulfil this objective, two mixed materials were prepared using a solid / solid mixture between laterite soil and granular ferric hydroxide for arsenic adsorption. Chemical analysis of laterite soil indicated a high amount of iron, aluminum and silicon. Batch experiments were conducted for As(V) adsorption using aqueous solutions. Results showed that the adsorption of arsenic (V) was strongly influenced by contact time, initial pH, adsorbent amount and initial As(V) concentration requiring their optimization. Indeed, the increase of the contact time between 5 and 90 min involved an increase of adsorption capacity up to 49.47 µg/g while a change of initial pH caused a variation of adsorption capacity from 49 to 42.38 µg/g. An increase of initial arsenic concentration showed a proportional increase of adsorption capacity for both mixed material while this capacity decreased when the adsorbent amount increased. Using both kinetic models, As(V) adsorption followed best the pseudo-second order kinetic.

Keywords: Adsorption, Arsenic, Granular ferric hydroxide, Laterite, Mixed materials

1 Introduction

Access to drinking water is one of the major challenges in Sub-Saharan Africa. The poor distribution of rainfall in space and time cause a lack of sustainability of surface water. At this regard, groundwater is the main source of fresh water supply in rural areas. However, ground waters often contains heavy metals and toxic metalloids such as arsenic, thus affecting their quality [1, 2, 3]. In Burkina Faso, excepted cyanides and mercury, arsenic is the pollutant that poses more health problem [4]. Groundwater contaminated with geogenic Arsenic is widely used as drinking water in West African countries. In Burkina Faso, ~560,000 people in rural communities are estimated to rely on water from arsenic-contaminated tube wells [3]. In recent decades, there has been a great deal of interest in research into the presence and behavior of arsenic in the environment due to the serious health problems caused by this metalloid [5, 6]. A study carried out in Yatenga province showed health problems such as cancers (skin, liver, lungs, bladder and kidneys) and dermatological effects (melanosis, keratosis) as typical signs of chronic exposure to arsenic [1, 2].

In order to reduce the arsenic concentration up to allowable value in drinking water, treatment methods such as filtration, coagulation-precipitation, reverse osmosis, electro-coagulation, adsorption, and coupled techniques have been developed to remove arsenic forms and species from water [7, 8]. However, they are either costly or complex, requiring skilled man power.

Nowadays, adsorption is considered as a suitable removal technology, particularly for developing regions, because of its simple operation, potential for regeneration, and little toxic sludge generation [9]. Processes based on the use of natural, locally available adsorbents are considered to be more accessible for developing countries, have a lower investment cost and a lower environmental impact (CO₂ emission). Laterite soils and iron oxyhydroxides have been widely used in the treatment of arsenic water using column and batch experiments [10, 11]. Among the iron oxyhydroxides, granular ferric hydroxide (GFH) commonly referred to as akaganeite has been the most effective in arsenic removal, but its cost and low regeneration after use do not allow its large scale use in developing countries [10, 11, 12]. GFH is a granular form of akaganeite (β-FeOOH) (grain sizes 0.2–2 mm) with an iron content of 610 g/kg and a specific surface area of 300 m²/g [13].

Laterite soils can be a promising alternative for the treatment of arsenic water because of their accessibility, their availability and their satisfactory adsorption capacity of arsenic (III) and (V) in many countries such as India, Vietnam, Bangladesh, Ghana [14-17] and Burkina Faso [4, 18]. Previous study showed the possible use of laterite in Burkina Faso for removing arsenic, but its removal capacity does not allow to have admissible value in drinking water [18]. In order to increase their performance, we propose to make a laterite-GFH mixture for arsenic adsorption. The influence of the operating parameters will be evaluated under various conditions and the kinetics of the process will be studied.

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2 Material and Methods

2.1 Preparation of two mixed materials

Laterite used in this work was collected at Balkuy district (12°17'23.35'' N, 1°27'46.90'' W) in Ouagadougou (Burkina Faso). The sampled lateritic crusts were transported to the laboratory and crushed manually with a hammer. Laterite soils were previously investigated and the results revealed the notable presence of iron oxides (hematite and goethite), aluminum oxide and quartz [18, 19]. Granular ferric hydroxide (US Filter/Wasserchemie GmbH) was commercially obtained from GEH Wasserchemie manufacturers in Germany. This material is predominantly akaganeite, a specific form of an iron oxide mineral and characterized by its crystalline structure, large specific surface area, and high porosity [20]. The important characteristics of GFH are summarized in Table 1.

For the preparation of mixed materials, the alternating sweeping method was used with a solid / solid mixture (m/m). The first material (noted M1) was prepared by making a mixture of GFH and laterite with a mass proportion $\frac{GFH}{Laterite} = \frac{2}{3}$. The second material (denoted M2) was a proportion $\frac{GFH}{Laterite} = \frac{3}{2}$. More the particle size is lower, the treatment is more effective [2]. The selection of grains size was carried out using two sieves of 0.25 mm and 1.25 mm pores.

2.2 Chemical characterization

Elemental and chemical compositions of laterite soil was determined using flame atomic absorption spectrophotometry (FAAS). Experimentally, 1 g of laterite was introduced into a beaker containing an acid solution (75 mL of 37% HCl + 25 mL of 65% HNO₃) and the mixture was stirred at 90 °C for 1 h. After stirring, the mixture was filtered and the concentrations of different chemical elements were determined by FAAS analysis using their different wavelengths.

2.3 Determination of pH at point of zero charge

The pH at point of zero charge (pH_{PZC}) is the pH at which the overall electrical charge of the adsorbent is neutral. It is determined according to the method of Noh *et al.* [21]. The procedure consists of putting different weights of laterite (2 g, 4 g and 5 g) in three Erlenmeyer flasks containing 10 mL, 20 mL and 25 mL of 0.1 M NaCl, respectively. Then, mixtures were stirring at a stirring speed of 170 rpm for 14 h at 26±2°C. After filtration, the pH of each solution was measured and the average pH value was calculated. This value indicates the pH at equilibrium corresponding to the pH at the point of zero charge of the material.

2.4 Determination of bulk density

The density is a physical parameter which makes it possible to estimate the mineralogical composition of raw material. It is the ratio of the material mass to the mass of the same volume of solvent used. The density (ρ) was determined by the following relation:

$$\rho = \frac{m_3 - m_1}{(m_2 - m_1) - (m_4 - m_3)} \quad (1)$$

where m_1 is mass of the empty flask and m_2 is mass of the flask filled with water; m_3 is mass of the flask containing 5 g of laterite;

and m_4 is mass of the flask containing 5 g of laterite supplemented with distilled water up to the mark.

2.5 Determination of the residual porosity

It was determined using the method of Batakou *et al.* [22] which consists to load the column with the adsorbent and set a flow rate of water at the inlet of the column. After, we determine the flow rate F_0 at the outlet of the column after 5 min and the flow rate F_1 at the outlet of the column after 1 hour of stirring. Porosity was calculated using the formula:

$$Porosity = \frac{F_1}{F_0} \times 100 \quad (2)$$

2.6 Equilibrium study

Batch experiments were carried out for arsenic removal in aqueous solutions with mixed materials M1 and M2. As (V) solutions are prepared by diluting an arsenic acid (H₃AsO₄) solution 1000 mg/L with distilled water. The operating parameters such as contact time, initial arsenic concentration, initial pH and adsorbent amount were varied and their influence on the adsorption capacity was studied. Experiments were carried out at the room temperature (26 ± 2°C) and repeated three times. Average values of data were used for calculations and figures. 50 mL of As(V) solutions of known concentration were added to 0.5 g of M1 or M2 and were stirred in 50 mL Erlenmeyer at 400 rpm throughout pH ranged from 3 to 11 at 26±2°C. Effect of initial arsenic concentration on arsenic adsorption capacity was studied between 0.2 and 1.25 mg/L using 0.5 g of M1 or M2 into 50 mL of As(V) solution with pH 3.6 -7, during 90 min of contact and 26±2°C at 400 rpm. Adsorbent amount was varied between 2 and 20 g/L using a mass from 0.1 g to 1 g of M1 or M2. After the filtration of mixture using Whatman filter, arsenic concentration in solutions was analyzed using Inductively Coupled Plasma-Optic Emission Spectrophotometry (ICP-OES). The spectrophotometer (Perkin Elmer, Optima 8000) was calibrated by using As (V) standard solutions of concentrations from 1 to 50 µg/L with a detection limit of 1 µg/L. Each experiment was carried out three times and the average value was calculated. The adsorption capacity (Q_e) expressed in µg/g was obtained from following relation:

$$Q = \frac{(C_0 - C_e)}{m} * V \quad (3)$$

where V represents the volume of the solution (L) and m is the weight or mass of adsorbent. C_0 and C_e representing initial and final concentrations of arsenic in solution (µg/L), respectively.

2.7 Kinetic study

The contact time at the saturation was determined by varying the contact time from 5 to 120 min with 0.1 g of adsorbent, pH 7, T=26±2 °C and 25 mL of solution containing initially an arsenic concentration of 1000 µg/L at 4000 rpm. Final concentration and its corresponding adsorption capacity were used to apply the kinetic models of pseudo-first order and pseudo-second order. In addition, the model of the interparticle diffusion was applied in order to determine the controlling step of adsorption kinetic. The pseudo-first order from equation of Lagergren and Svenska [23] based on the adsorption capacity (Eq. 4) and pseudo-second order from equation of Ho and McKay [24] based on the equilibrium

between adsorbate and adsorbent (Eq. 5) were used for kinetic modelling.

$$\ln(Qe - Qt) = -k_1 t + \ln Qe \quad (4)$$

$$\frac{t}{Qt} = \frac{1}{Qe} t + \frac{1}{K_2 Qe^2} \quad (5)$$

Plots $\ln(Qe - Qt) = f(t)$ and $t/Qt = f(t)$ are drawn and the values of slope and intercept will be used for kinetic modelling. The rate limiting step of the sorption can be qualitatively determined by analyzing kinetic data using Weber-Morris' model [25, 26]:

$$Qt = k_1 t^{0.5} + C \quad (6)$$

where, k_1 is the diffusion coefficient ($\mu\text{g/g.min}^{0.5}$) and C is a constant that gives an indication of the thickness of boundary layer.

3 Results and discussion

3.1 Chemical and physico-chemical composition

All the characteristics of GFH and laterite are given in Table 1. Properties of GFH were determined in previous work by Amy *et al.* [20]. Granular Ferric hydroxide called akaganeite is a ferric hydroxide crystallized under $\beta\text{-FeOOH}$ form and contains chloride, ferric trihydroxide $\text{Fe}(\text{OH})_3$ and $\alpha\text{-FeOOH}$ form. A rapid comparison indicates that GFH has high surface area and pH_{ZPC} indicating its high efficiency compared to laterite soil. However, high porosity of laterite would indicate the high pore volume and radius. High density of laterite is correct because it is more heavy compared to GFH which breaks during stirring of the solution in the beaker. Table 2 shows the elemental and chemical composition of raw laterite using SAAF analysis. We notice the strong presence of iron, aluminum and silicon while a few quantity of manganese, calcium, magnesium, copper and zinc was observed. These chemical elements were comparable to those commonly identified in laterite soils [17-19].

3.2 Influence of operating parameters on arsenic (V) removal

3.2.1 Effect of contact time

Figure 1 indicates a rapid initial uptake rate of arsenic, followed by a slower removal that gradually approaches an equilibrium condition from 5 min to 90 min. We noticed the increase in adsorption capacity when the stirring time increases according to three phases: A first rapid increase between 5 and 15 minutes with a better efficiency of M1 was due to the high presence of active sites for adsorption of arsenates. The slower adsorption with M2 was likely because of the decrease in the driving concentration difference between the bulk solution and the surface, which decelerates transport of the arsenic species to

the GFH surface. The second step between 15 and 60 minutes with better efficiency of M2 shows a slight increase of adsorption capacity due to the decrease of active sites when the contact time increases.

Table 1: Physical-chemical characteristics of Laterite and GFH

Property	Quantitative value	
	Laterite	GFH
pH_{ZPC}	7.19	7.6 – 7.8
Surface area	42.39	240 - 300
Grain size (mm)	0.25 - 1.25	0.32 - 2
Bulk density (g/mL)	2.6	1,19
Moisture content (%)	ND	43 - 48
Residual porosity (%)	91.62	72 - 77

ND: Not determined

The performance of M2 can be explained by the high content of iron oxide and its affinity with arsenic favoring As(V) adsorption. The last relatively constant phase from 60 up to 120 minutes with a better efficiency of M2 indicated a possible saturation of surface area of mixed materials and their active pore sites. Analysis of data in Table 2 shows a high quantity of hematite, quartz and alumina in laterite. In addition, it was noted a low presence of oxides from manganese, calcium, magnesium, copper and zinc oxides in the laterite composition. High presence of iron oxide should be responsible of high adsorption of arsenic regarding the affinity arsenic-iron. Literature data revealed that more the material contains iron oxide, more it is efficient in arsenic adsorption (xxx, xxx). However, the adsorption capacity of an adsorbent is influenced by operating conditions such as contact time, pH of the solution, initial arsenic concentration and adsorbent amount. This result is attributable the driving force which gradually decreases over time, while the agitation remains constant leading the system to equilibrium where no further withdrawal is possible with increasing time [27, 28]. The material M2 was more efficient because its performance continues to increase until equilibrium was reached after 90 minutes.

3.2.2 Effect of initial pH

The pH is one of the important factors which significantly affects the adsorption of arsenic in aqueous solutions as it modifies the surface charge of adsorbent [29].

Table 2: Chemical and elemental composition of raw laterite (m/m)

Element	Fe	Al	Si	Mn	Mg	Ca	Cu	Zn
Elemental composition (%)	15.06	8.20	11.88	0.26	1.05	2.52	< 0.1	< 0.1
Oxide	Fe_2O_3	Al_2O_3	SiO_2	MnO	MgO	CaO	CuO	ZnO
Chemical composition (%)	31.75	22.82	37.46	0.48	2.25	5.18	< 0.1	< 0.1

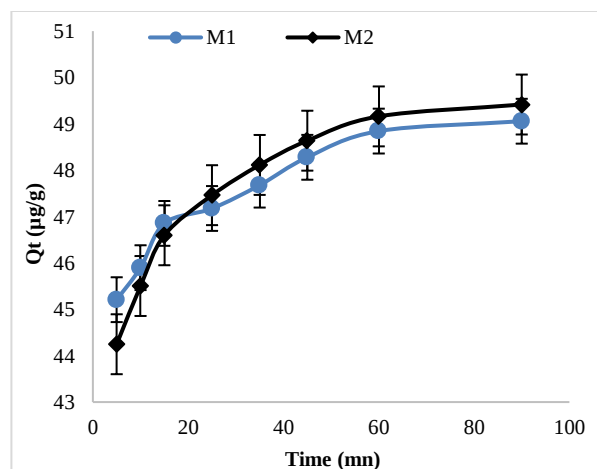


Figure 1: Effect of contact time on As(V) adsorption capacity using M1 and M2 with $C_0 = 0.5$ mg/L, $V = 50$ mL, $m = 0.5$ g, $pH = 3.6$ and $T = 25^\circ\text{C}$

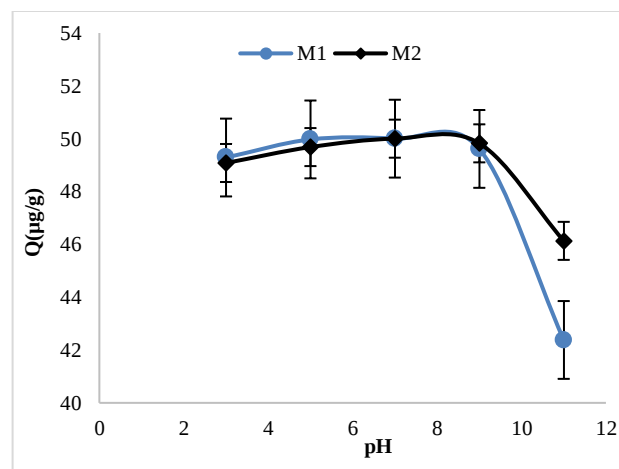


Figure 2: Effect of initial pH on As(V) adsorption capacity using M1 and M2 with $C_0 = 0.5$ mg/L, $t = 90$ min, $m = 0.5$ g, $V = 50$ mL and $T = 25^\circ\text{C}$

Figure 2 shows the influence of pH on the adsorption capacity using materials M1 and M2. Results showed the arsenic adsorption capacity (V) as a function of pH is presented in different stages. The first phase between pH 3 and 8 indicates a slight increase of adsorption capacity using M1 and M2 with better efficiency using M1. Indeed, for pH below the isoelectric point, the density of positive charges on the surfaces of M1 and M2 promotes the binding of anionic forms of arsenic. Between pH 7 and 8, the adsorption capacity is the same and maximum for M1 and M2 and remains relatively constant. This result can be explained by a decrease in the charge density on materials M1 and M2 whose surfaces are almost neutral because the pH values are closed to the isoelectric points of M1 and M2. Above $pH = 8$, adsorption capacities decreased quickly using both two materials with a better efficiency of M2. This quick decrease could be explained the negatively charged surfaces of two materials which do not promote the adsorption of arsenates ($H_2AsO_4^-$ and $HAsO_4^{2-}$) at pH above the isoelectric points of M1 and M2. This decrease in adsorption capacity should be due to a repulsion of similar charges of arsenates and the surface on the one hand, and, on the other hand, between arsenates and hydroxyl ions [4, 30]. Adsorption of arsenic (V) species, such as $H_2AsO_4^-$ and $HAsO_4^{2-}$, onto hydrous iron oxide is known to take place via Coulombic as well as Lewis acid-base interactions (ligand exchange reactions) and to form monodentate and bidentate inner sphere complexes.

3.2.3 Effect of initial As(V) concentration

Figure 3 shows the influence of initial arsenic concentration on the adsorption capacity. Results indicated a proportional increase of adsorption capacity using M1 and M2 when the initial concentration of As (V) increased. This could be explained by the accessibility of a greater number of active sites by As (V) species at high concentrations of arsenic [4]. In addition, the more the initial arsenic content increases, the more the number of arsenate ions increases and the available active sites are filled. The collinear line obtained using M1 and M2 shows that both two materials have the same adsorption capacity for arsenic by increasing the initial arsenic concentration.

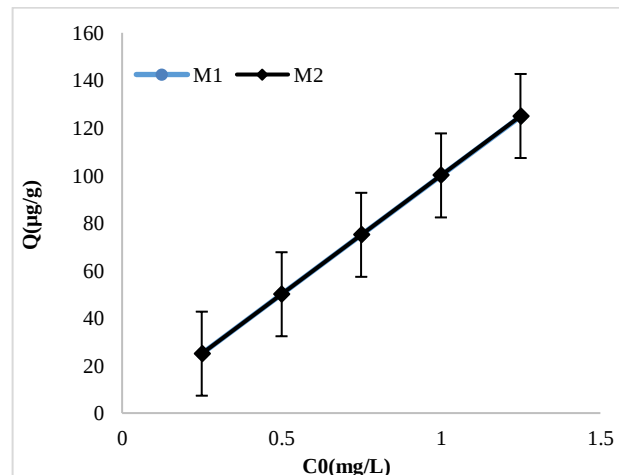


Figure 3: Effect of initial As(V) concentration on its adsorption capacity using M1 and M2 with $m = 0.5$ g, $V = 50$ mL, $pH = 3.6 - 7$, $t = 90$ min and $T = 25^\circ\text{C}$

3.2.4 Effect of adsorbent amount

Figure 4 shows the effect of adsorbent amount on As (V) adsorption at natural pH and room temperature. In figure 4, we notice that arsenic (V) adsorption capacity decreases with the increase of adsorbent dose using two materials M1 and M2. However, As(V) removal percentage increased from 96% to 99.5% when the adsorbent mass increased in the same range between 0.1 g and 1 g. This result could be explained the increase of active sites due to the increase of adsorbent mass and their availability for adsorption [31, 32].

3.3 Kinetic modelling

In order to better elucidate the kinetics of arsenic (V) adsorption on M1 and M2 materials, the equation of Lagergren *et al.* [23] and that of Ho and Mckay [24] describing the pseudo-first order and pseudo-second order kinetics are applied. Results are given in Figures 5 and 6. Values of kinetic constants are listed in Table 3. Table 3 gives the kinetic parameters (Q_{exp} , Q_{theo} , and k)

and the correlation coefficient R^2 in As (V) adsorption of using M1 and M2 for each kinetic model.

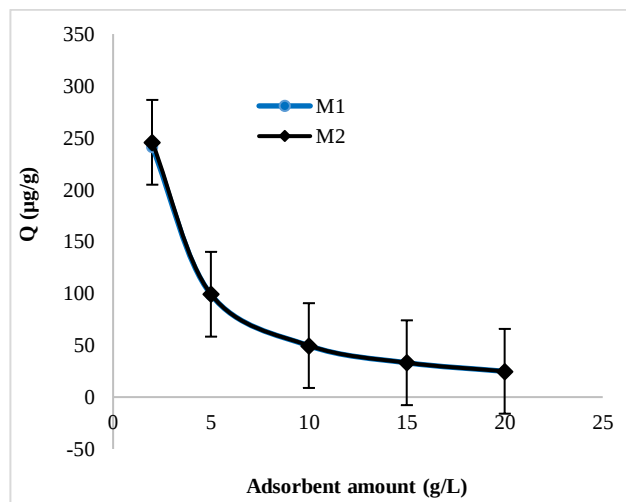


Figure 4: Effect of adsorbent amount on As(V) adsorption capacity using M1 and M2 with $C_0 = 0.5$ mg/L, pH = 3.6, $t = 90$ min and $T = 25^\circ\text{C}$

Rapid comparison shows that the theoretical (Q_{theo}) value is closed to experimental (Q_{exp}) value for the pseudo-second order kinetic model using both two materials. In addition, values of the correlation coefficients (R^2) for pseudo-second order kinetic model using M1 and M2 are higher compared to the pseudo-first order model. These results indicate that the kinetic of As (V) adsorption onto M1 and M2 would be more explainable by pseudo-second order suggesting the existence of chemisorption (exchange of ligands) of arsenic (V) [7]. High correlation coefficient R^2 using M2 could be explained by high iron oxides in this material favoring the adsorption of As(V). It is generally known that the sorption process is a rate-controlled process, in which the slowest step determines the process rate limiting step. Kinetic data were further analyzed assuming that the mechanism of As(V) sorption can generally be described by four consecutive rate controlling steps, which are external mass transfer, film diffusion, intraparticle diffusion, and surface interactions on active sites [33, 34]. The sorption process is said to be intraparticle diffusion controlled, if the straight line plot passes through the origin, while the boundary layer diffusion (external mass transfer or film diffusion) may take place, if it does not pass through the origin [33]. Figure 7 indicates the plot representing this kinetic model in As(V) adsorption. Calculations revealed the correlation coefficient value was of 0.91, and the constant k_1 of $0.71 \mu\text{g/g.min}$. The linear form of curve doesn't pass through the

origin, indicating that the intraparticle diffusion is not the limiting step of the process and the mechanism of the process is complex. However, the high value of intercept C (43.44) indicates the effect of transboundary layer. We conclude that the process of As(V) adsorption is controlled by the film diffusion as described in the literature [33, 35, 36].

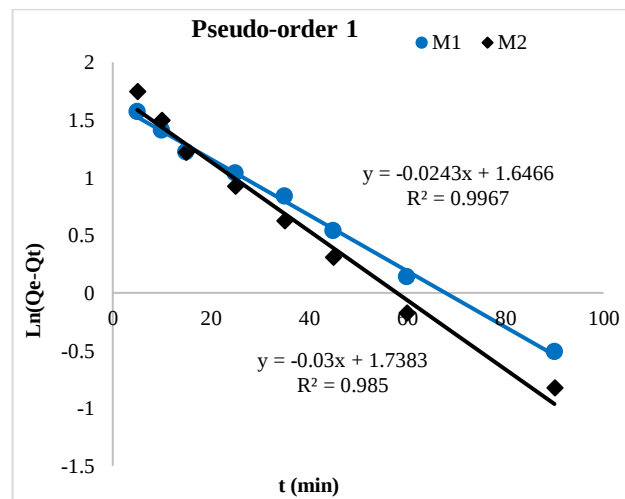


Figure 5: Representation of pseudo-first order model in As(V) adsorption

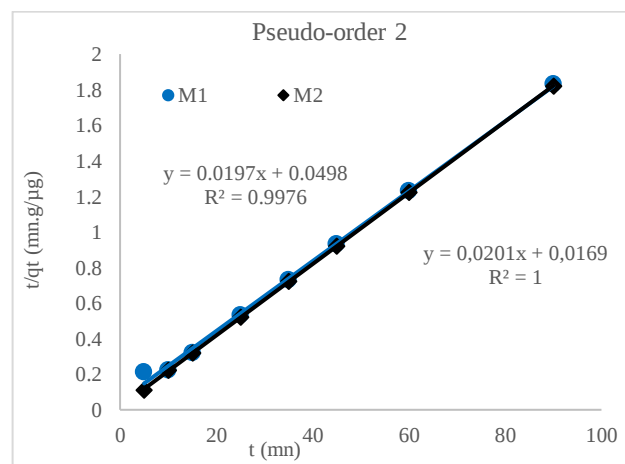


Figure 6: Representation of pseudo-second order model in As(V) adsorption

Table 3: Kinetic parameters of two models for arsenic adsorption using M₁ and M₂

	M ₁				M ₂			
	K_1 (min ⁻¹)	Q_{theo} (mg/g)	Q_{exp} (mg/g)	R^2	K_2 (min ⁻¹)	Q_{theo} (mg/g)	Q_{exp} (mg/g)	R^2
Pseudo-first order model	0.05	0.05	0.049	0.99	0.05	0.05	0.05	0.98
	M ₁				M ₂			
	K_2 (g.mg ⁻¹ .min ⁻¹)	Q_{theo} (mg/g)	Q_{exp} (mg/g)	R^2	K_2 (g.mg ⁻¹ .min ⁻¹)	Q_{theo} (mg/g)	Q_{exp} (mg/g)	R^2
Pseudo-second order model	26.82	0.05	0.049	0.99	22.33	0.05	0.05	1

Table 4: Comparison of adsorption capacity of M1 and M2 with literature data

Adsorbent	pH	Surface area (m ² /g)	Temperature (°C)	Adsorption capacity (mg/g)		References
				As (III)	As (V)	
Granular ferric hydroxide	3	240	26±2	-	0.10	[11]
GFH	7	-	-	8.5		[37]
Laterite soil	7.2	18.05	29	0.17	0.1	[11]
GFH mixed laterite	3.6	-	26 ± 2	-	0.049	Present study (M1)
	3.6	-	26 ± 2	-	0.05	Present study (M2)
Ferric ion loaded red mud	7	-	25	4	-	[38]
Ferrihydrite	6	141	27	-	0.025	[39]

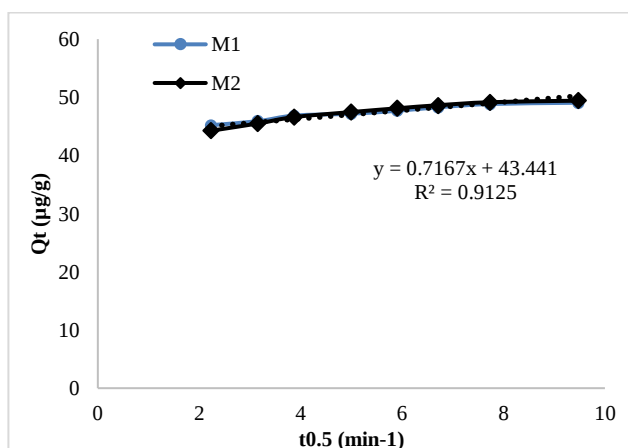


Figure 7: Intraparticle diffusion model for As(V) adsorption using M1 and M2

3.4 Comparison of adsorption capacity

Other mixed materials have been prepared and used for arsenic adsorption in literature [37, 38, 39]. To confirm our study, adsorption capacities of M1 and M2 were compared to that of other adsorbents with the challenge of the differences in experimental conditions which impact considerably on the adsorption capacities. However, for the sake of comparison, the values of monolayer adsorption capacity (q_m) collected from the literature for various adsorbents under the similar conditions and those of M1 and M2 are listed in Table 4. The comparison of adsorption capacities (Table 4) indicated that M1 and M2 have an adsorption capacity (0.05 mg/g) greater compared to the one of ferrihydrite and iron oxide coated sand used as adsorbents [39, 40]. Moreover, this capacity remains lower compared to GFH used alone and iron coated materials previously studied [11, 37, 38]. However, the comparison doesn't be much exacted because some operating conditions such as adsorbent mass, temperature pH of solution and surface area of adsorbent which cannot be the same.

3 Conclusion

This work was able to assess the performance of mixed materials for arsenic removal with an adsorption capacity of 0.05 mg/g. Batch experiments revealed the influence of contact time, initial pH, adsorbent amount and initial concentration of arsenic requiring their optimization for application in column experiments or using natural arsenic water. As(V) adsorption was

occurred according to pseudo-second order kinetic with film diffusion as controlling step of the process.

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Ethical issue

Author is aware of, and comply with, best practice in publication ethics specifically with regard to authorship (avoidance of guest authorship), dual submission, and manipulation of figures, competing interests and compliance with policies on research ethics. Authors adhere to publication requirements that submitted work is original and has not been published elsewhere in any language.

Competing interests

The author declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

Authors' contribution

Dr. Yacouba Sanou has been developed the original idea and the protocol, abstracted and analyzed data, wrote the manuscript. He has studied the concept and design of this work. He was involved in drafting the manuscript or revising it critically for important intellectual content; and he is the guarantor;

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