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Functionalized C,N-Bipyrazole Receptor Grafted onto Silica Surface for Arsenic Adsorption and Its Antibacterial Activity

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Abstract

The present study is based on the investigation of performance of C,N-bipyrazole receptor grafted onto silica surface (SG2P) of adsorption Arsenic (AS) from aqueous solutions. The effects of operating parameters that include pH, contact time, concentration of As and dosage of adsorbent on adsorption were accomplished. The results clearly showed that the removal efficiency of As was decreased with an increasing of As concentration, pH, and temperature, while it was continuously increasing with time and adsorbent dose. Moreover, the removal efficiency of Cr (VI) adsorption was 75% corresponding to pH; temperature (°C), initial concentration (ppm) and weight of dose (g) were 6, 25, and 0.04 respectively at 24 hours. The adsorption capacity of the synthesized sorbent (SG2P) for arsenic at pH < 7 from pseudo second order in batch experiments was efficient at 20 minutes. Furthermore, the results obtained in this study presented that the antimicrobial activity was limited where the Escherichia coli (ATCC25922) and Staphylococcus aureus (ATCC25932) were as a reference strains, while, the SG2P was able to inhibit growth only at high concentration (MIC = 1.5625 mg/ml).

Keywords

Antimicrobial Activity, Arsenic, Adsorption, Pseudo Second Order, *Escherichia coli*

1. Introduction

Arsenic (As) is the 20th most abundant element present in the earth's crust; it is

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considered as a toxic trace element present in natural waters (ground and surface water), and has become a major unavoidable threat for the life of human beings and useful microorganisms. Arsenic concentration in soils and water can become high due to several reasons like, mineral dissolution, use of arsenical pesticides, disposal of fly ash, mine drainage, and geothermal discharge [1]. The major arsenic species present in natural waters are arsenate ions: H_3AsO_4 , $H_2AsO_4^2$, $HAsO_4^2$, and AsO_4^3 (oxidation state V) and arsenite ions, $H_3AsO_3^3$, $H_2AsO_3^-$ and $HAsO_3^{2-}$ (oxidation state III). However, As(V) ions are most widespread in oxygenated water while As(III) is found in anaerobic conditions like in well water or in groundwater. The concentration of arsenic species is mainly dependent on redox potentials [2] and pH [3].

Recently, the environmental fate and behavior of arsenic (As) are receiving increased attention due to the arsenic (As) pollution in groundwater. It has been a serious health threat to the human beings in the world. Occurrence of arsenic in groundwater above the permissible value (>10 μ g/L WHO) is one of the wide spread problem owing to its toxicity and carcinogenicity [4]. In natural waters arsenic normally occurs in the oxidation states +III (arsenite) and +V (arsenate). The removal of As(III) is more difficult than the removal of As(V). Thus, As(III) has to be oxidized to As(V) prior to its removal. In this context, Arsenic is considered as a toxic metal which is toxic to living organisms and harmful to the environment when it is directly discharged into water body. Arsenic can be easily absorbed by fishes and vegetables due to its high solubility in the aquatic environment and may be accumulate in the human body by the means of food chain. So, it is indispensable to treat water and/or wastewater containing toxic metals like arsenic to remove their adverse effects on human and ecology. Recently several methods of treating water for arsenic reduction are being investigated; such as ion-exchange, adsorption, chemical precipitation, membrane filtration, and flocculation and electrochemical methods [5]. Adsorption is a fundamental process in the physicochemical treatment of municipal wastewaters, a treatment which can economically meet today's higher effluent standards and water reuse requirements. Also, it considered as a broad spectrum of physical, biological, and chemical processes and operations in the environmental field. Purification of gases by adsorption has played a major role in air pollution control, and adsorption of dissolved impurities from solution has been widely employed for water purification. Adsorption is now viewed as a superior method for wastewater treatment and water reclamation [6] [7]. Based on literature reviews, so far, no other studies adsorption on new chemical adsorbent (C,N-bipyrazole with a 3-glycidoxyprpoyl-trimethoxyilane silylant agent) onto a silica surface in short (SG2P) have used. The objectives of this study were to prepare supported SG2P and to test its performance for arsenic removal from water. The effects of various parameters such as adsorbent dosage, pH, contact time, concentration of As and temperature on arsenic removal were assessed.

2. Materials and Methods

2.1. Chemicals and Instruments

The entire chemicals used were of AR grade and were used without further purification. All aqueous solutions were prepared in double distilled water. The distilled water was inspected for arsenic concentration. The concentration was lower than 10 ppb, the detection limit that can be easily accomplished by the analytical methods used in this study [8]. The permissible limit of arsenic is 10 ppb. Stock solutions of arsenic were prepared in reverse deionized (DI) water using arsenic trioxide. Arsenic working solutions were freshly prepared by diluting arsenic stock solutions with DI water. H_2SO_4 and NaOH (0.1 N) were used for adjusting the pH of the arsenic solution. Then a calibration curve of absorbance versus concentration (**Figure 1**) was constructed at the time of analysis. Calibration coefficient (R^2) was at least 0.983 for all analyses. Appropriately diluted solutions were made from the stock as and when required for calibration purpose.

The instruments used for the experimental work are Electronic Balance, pH meter; Spectrophotometer, Gutzeit apparatus, incubator and Gyro shaker which were used from the college laboratory and all the glass that used were made up of Borosilicate. On the other hand, the amount of metal ion that was sorbed and the percent removal of metal ion by adsorbent were calculated by applying Equation (1) and Equation (2) respectively:

$$q = (Co - Cf) \div .V \tag{1}$$

$$\%Removal = (Co - Cf) \div Co \ 100 \tag{2}$$

where q is the amount of metal ion sorbed by the adsorbent (mg/g); Co is the initial metal ion concentration; (mg/L), Cf is the final ion concentration (mg/L) after the chemo-sorption occurred, V is the volume of aqueous solution (L) in contact with the adsorbent, and m is the mass (g) of adsorbent.

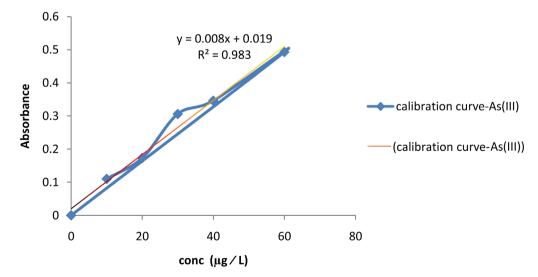


Figure 1. Linear calibration curve of absorbance vs. concentration for As (III) concentrations in the range $10 - 60 \mu g/L$.

2.2. Methods of Analysis

2.2.1. Preparation of Adsorbent Substance (SG2P)

After converting the hydroxy-bipyrazolic ligand 4 to the alcoolate derivative using sodium metal in tetrahydrofuran, the resulting salt (103 mol) was added to a suspension of 3-glycidoxypropyl-functionalized silica (SiPz) (1.00 g) in 30 ml of dimethylformamide (DMF). The mixture was stirred and refluxed under nitrogen for 24 h. The solid material was filtered, and the residue was washed with DMF, toluene, water (distilled and deionized), methanol, dichloromethane (150 ml of each) and finally dried [9] [10].

2.2.2. Determination of Optimum Dosage of Adsorbent

To determine the optimum dosage of SG2P, it was added to the conical flask in different dosage varying from (0.02 g, 0.04 g, and 0.1 g up to 0.15 g), containing 10 mL concentration of arsenic solution (10 μ g/L) at pH 6. The solution in the conical flask was subjected to stirring for optimum contact time, filtered and analyzed for residual arsenic concentration. The dosage which provides minimum residual concentration is chosen as optimum dosage.

2.2.3. Selection of Optimum Contact Time

The adsorption is strongly influenced by the contact time. To study the effect of contact time, 10 mL of 10 μ g/L arsenic solution was mixed with 0.02 g of adsorbant (SG2P), stirred at different contact times varying from (5 mins, 15 mins up-to 180 mins). Then filtrate was analyzed for arsenic concentration using uv visible spectrophotometer.

2.2.4. Selection of Optimum pH on Arsenic

It was reported in prior studies that pH is a central factor that affects the performance of adsorption process. The effect of pH on arsenic adsorption was studied by performing equilibrium adsorption tests at different initial pH values. *i.e.* from 1.0 to 12.0. The pH of solution was adjusted by using 0.1N $\rm H_2SO_4$ or 0.1N NaOH. The pH which gives minimum residual concentration is chosen as optimum pH.

2.2.5. Determination of Optimum Temperature on Arsenic

The effect of temperature was investigated at different degrees, 25° C, 35° C, 45° C and 55° C. For each temperature, a 0.02 g adsorbent (SG2P) samples was added to 10 mL of As(III) solutions with concentration 10 µg/L at pH around 6. The temperature which gives minimum residual concentration is chosen as optimum temperature.

2.2.6. Selection of Optimum Initial Concentration of Arsenic

Al of these concentrations of 10, 20, 30, 40 and 60 ppb solutions were prepared from the stock solution to find out the optimum concentration, 0.02 g of adsorbent (SG2P) was added to a number of tubes contains 10 ml of the previous concentrations of As(III) solutions under optimized temperature (25°C) and pH 6 for 30 min, the absorbance of the solution above the solid residue was meas-

ured by Flame Atomic Absorption at 193.7 nm.

2.2.7. Determination of Antibacterial Activity of Adsorbent Substance (SG2P)

Antibacterial activity of SG2P was determined by broth micro-dilution method in which SG2P was serially diluted and examined against the bacteria. Minimum inhibitory concentration (MIC) was considered the lowest concentration of substance that inhibited visible growth of bacteria.

3. Results and Discussions

3.1. Effect of Contact Time

Effect of contact time was investigated and graph of percentage of arsenic removal versus time in minutes was schemed as shown in **Figure 2**. From the graph it is clear that the adsorption curves are characterized by sharp rise in removal efficiency of Arsenicat the first 20 minutes. Then it has negligible effect. This is may be due to accumulate of chromium ions at the surface of As(III). In this context, the optimum time for the adsorption of arsenic on SG2P was 20 minutes.

3.2. Effect of Adsorbent Dosage

Effect of adsorbent dosage was examined and graph of percentage of arsenic removal versus dosage was plotted as shown in **Figure 3**. From the graph it is observed that, as the dosage of SG2P increases, amount of arsenic decreases, sharply in the beginning and attains maximum later. This is mainly due to the large contact surface area and adsorption sites available for adsorption. The dosage, at which maximum removal is attained, is taken as maximum dosage. The removal efficiency on arsenic was found to be 77%, 80.5%, and 81.5% with an optimum dosage of 0.05 g, 0.1 g, and 0.15 g respectively.

3.3. Effect of pH

The adsorption of arsenic from aqueous solution using GS2P in this research was found to be a lower pH (pH 3) dependent process. The graph of percentage of arsenic removal versus pH was plotted as shown in **Figure 4**. From the graph it is observed that, arsenic is removed more effectively in slight acidic range.

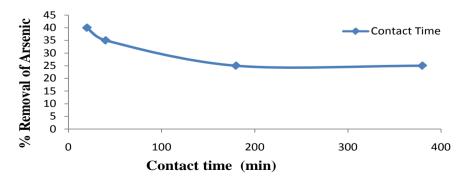


Figure 2. Effect of contact time.

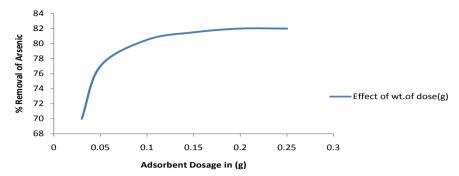


Figure 3. Effect of adsorbent dosage.

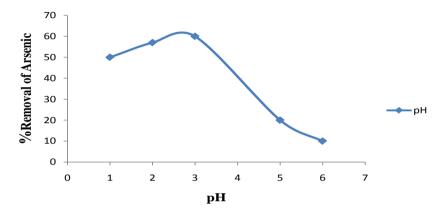


Figure 4. pH effect on As(III) adsorption ($C_o = 10 \mu g/L$, T = 25°C, adsorbent dose = 0.02 g).

This is because the solution contains protons that bind with electrons from oxygen, so the negative charge on the surface attract the positive ions of As, where at high pH in alkaline conditions the result showed is not favorable for arsenic sorption because carboxyl, hydroxyl, and amide groups of the adsorbent become negatively charged and a high density of OH⁻ at alkaline conditions would compete with all anionic species of As(III) [11].

3.4. Effect of Temperature

Effect of temperature was investigated and graph of percentage of arsenic removal versus temperature was plotted as shown in **Figure 5**. From the graph it is observed that, arsenic is removed more effectively in high temperature range. As expected, increasing temperature will lead to increase in temperature results in increase in mobility of large metal ions, which indicates that the adsorption process is endothermic, leads to swelling effect within the internal structure of adsorbent that in turn, encourages the metal ions to penetrate further and increase in the kinetic energy of adsorbent particles that increase the collision between adsorbent and adsorbate molecules results in enhancing the removal of metals from adsorbent surface [12].

3.5. Effect of Arsenic Concentration

The effect of arsenic concentration on the adsorption was studied under optimized

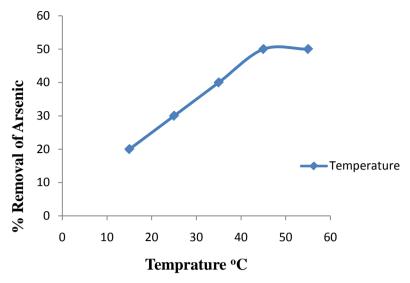


Figure 5. Effect of temperature on As(III) adsorption ($C_o = 10 \text{ mg/L}$, time = 60 min, adsorbent dose = 0.02 gm).

pH found from our previous results. Concentrations of Arsenic varied from 10 - 40 ppb. 10 ml of each concentration of arsenic was treated with the adsorbent at pH value of 3.0. The results are illustrated in **Figure 6**, which show GS2P is more efficient adsorbent at low concentrations of arsenic. Because at the beginning the removal of As(III) ions is high due to large surface area of adsorbent and when the saturation point is reached, at equilibrium the capacity of the (SG2P) to adsorb As(III) ions is controlled and the ions transported from the exterior to the interior sites of the (SG2P) particles [13].

3.6. Adsorption Isotherm of Arsenic

Adsorption isotherms are used to explain the equilibrium of metal ions that occurs between the solid phase of adsorbent and the aqueous solution. Freundlich and Langmuir isotherms are the most models that widely used to determine some kinetic and thermodynamic parameters that can give a clearer image about the binding mechanism.

3.6.1. Freundlich Adsorption Isotherm

The Freundlich isotherm is an empirical equation employed to describe heterogeneous systems. This model is specified with the following equation [14]:

$$Q_a = K_E C_a^{1/n} \tag{5}$$

The linear form of this equation can be written as:

$$\ln Q_e = \frac{1}{n} \ln C_e + \ln K_F \tag{6}$$

where, K_F and n are Freundlich constants with K_F is an approximate indicator of adsorption capacity of the sorbent and n giving an indication of how favorable the adsorption process. The magnitude of the exponent, 1/n, gives an indication of the favorability of adsorption. If value of 1/n is below one, it indicates a normal

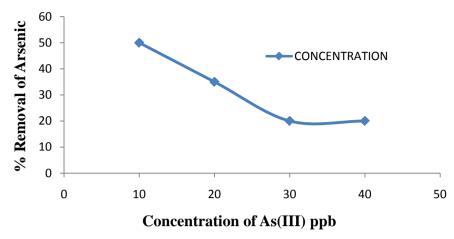


Figure 6. Effect of As(III) concentration on adsorption.

adsorption. If n lies between one and ten, this indicates a favorable sorption process [15]. To determine the constants K_F and n, the linear form of the equation may be used to produce a graph of $\ln(Q_e)$ against $\ln(C_e)$ as shown in **Figure** 7.

From Table 1 the values of n = 1.6949 and 1/n = 0.59, as Freundlich isotherm model, these values indicate that the sorption of As(III) on SG2P is favorable and the R^2 value is 0.911.

3.6.2. Langmuir Adsorption Isotherm

This describes quantitatively the formation of a monolayer adsorbate on the outer surface of the adsorbent, and after that no further adsorption takes place. Thereby, the Langmuir represents the equilibrium distribution of MB dye between the solid and liquid phases. The Langmuir isotherm is valid for monolayer adsorption onto a surface containing a finite number of identical sites. The model assumes uniform energies of adsorption onto the surface and no transmigration of adsorbate in the plane of the surface. Based upon these assumptions, Langmuir represented the following equation [14] [16]:

$$\frac{C_e}{Q_e} = \frac{1}{Q_m} C_e + \frac{1}{Q_m K_L} \tag{7}$$

where:

 C_e = the equilibrium concentration of adsorbate (mg/L);

 Q_e = the amount of As(III) adsorbed per gram of the adsorbent (mg/g);

 Q_m = maximum monolayer coverage capacity (mg/g);

 K_L = Langmuir isotherm constant (L/mg).

The values of Q_m and K_L were computed from the slope and intercept of the Langmuir plot of C_e/Q_e versus C_e . From Langmuir plots which is shown in **Figure 8**. amount adsorbed for monolayer formation (Q_m) , Langmuir adsorption-desorption equilibrium constant (K_L) and regression constant (R^2) were determined and values are shown in **Table 2**.

The essential characteristics of the Langmuir isotherm can be expressed in

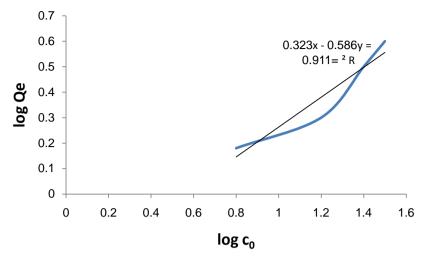


Figure 7. Freundlich plot for As(III) adsorption on (SG2P).

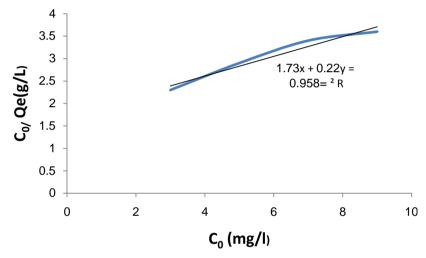


Figure 8. Langmuir plot for As(III) adsorption on (SG2P).

Table 1. Parameters and correlation coefficient of Freundlich isotherm model for adsorption of As(III) onto SG2P.

Freundlich isotherm model parameters									
	Parameters								
Adsorbate	$\frac{1}{n}$	n	$K_F = (\text{mg/g})$	R^2					
SG2P	0.59	1.6949	2.1	0.911					

Table 2. Parameters and correlation coefficient of Langmuir isotherm model for adsorption of As(III) onto (SG2P).

Langmuir isotherm model parameters								
Adsorbate	Parameters							
	$Q_m (\text{mg/g})$	$K_L = (L/mg)$	R_L	R^2				
SG2P	4.61	0.578	0.166	0.958				

terms of a dimensionless constant separation factor R_L that is given by the following equation [15]:

$$R_L = \frac{1}{\left(1 + K_L C_o\right)} \tag{8}$$

where, C_0 is the highest initial concentration of adsorbate (mg/L).

The value of R_L indicates the shape of the isotherm to be either unfavorable $(R_L > 1)$, linear $(R_L = 1)$, favorable $(0 < R_L < 1)$, or irreversible $(R_L = 0)$. The R_L values between 0 and 1 indicate favorable adsorption. The value of R_L in the present investigation was found to be 0.166 at 25 °C indicating that the adsorption of As(III) dye on (SG2P) is favorable [17].

3.7. Rate Order of the Adsorption Process

Uptake of each metal ion from different initial concentrations of the metal can be used to study the dependency of the rate of adsorption on the concentration of metal ion left in solution. Thus, the reaction order of the adsorption process can be determined. Pseudo first-order equation was applied for evaluation the adsorption kinetics for As(III) onto SG2P, the rate constant for the adsorption K_1 was evaluated. The pseudo first-order equation expressed as Equation (4):

$$\log\left(Q_e - Q_t\right) = \log Q_e - \left(\frac{K_1}{2.303}\right)t\tag{9}$$

where Q_e is the adsorption capacity of the SG2P at equilibrium (mg/g), Q_t is the amount of As(III) adsorbed at time t (mg/g) and K_1 is the pseudo first order rate constant (min⁻¹).

A linear plot of $\log(Q_e - Q_t)$ against time allows obtaining the rate constant (**Figure 9**). If the plot was found to be linear with good correlation coefficient, indicating that Lagergren's equation is appropriate to As(III) sorption on SG2P so the adsorption process is a pseudo first order process. The Lagergren's first order rate constant (K_1) and Q_e determined from the model are presented in **Table 3** along with the corresponding correlation coefficients. It was observed that the pseudo first order model did not fit well. It was found that the calculated Q_e values do not agree with the experimental Q_e value This suggests that the adsorption of As(III) does not follow first-order kinetics.

From **Figure 9**, the R^2 value shows that the adsorption As(III) onto SG2P does not obey the pseudo first-order kinetics, and the pseudo first-order model is not appropriate for the adsorption.

The Lagergren pseudo first-order rate constant (K_1) and (Q_e) values are represented in **Table 3**.

The pseudo second order kinetics may be expressed in a linear form as integrated second order rate law [13]:

$$\frac{t}{Q_{t}} = \frac{1}{Q_{e}}t + \frac{1}{K_{2}Q_{e}^{2}} \tag{10}$$

where K_2 is the pseudo second order rate constant (g·mg⁻¹·min⁻¹).

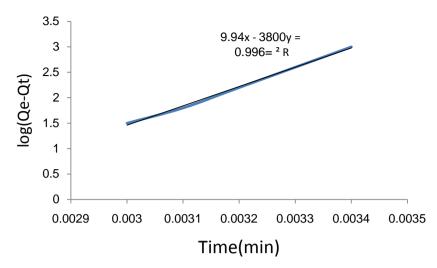


Figure 9. Pseudo first-order adsorption kinetics of As(III) onto (SG2P).

Table 3. Pseudo first-order parameters for As(III) adsorption onto SG2P at 25°C.

	0 ()		pseudo first-order	
Adsorbent	Q_e (exp) (mg/g)	K_1 min ⁻¹	Q_e (calculated) (mg/g)	R^2
SG2P	2.3	4.6×10^{-s}	0.51	0.408

Pseudo second-order adsorption model for As(III) adsorption onto SG2P was applied and the rate constant for the adsorption K_2 was evaluated as shown in Figure 10.

The results obtained shows that the value of linear regression coefficient R^2 is 0.999, the values of Q_e experimental, K_2 , R^2 and Q_e calculated were listed in **Table 4**.

The results shows that the Pseudo second-order kinetic model perfect fit with experimental data and the value of $R^2 = 0.9998$, by comparing (Q_e) experimental and (Q_e) calculated values from **Table 4**, there is agreement between experimental and calculated values for pseudo second order model, and K_2 value greater than K_1 value, so the pseudo second order model is favorable model for As(III) adsorption onto SG2P.

3.8. Adsorption Thermodynamics

Adsorption thermodynamics were determined using the thermodynamic equilibrium coefficients obtained at different temperatures and concentrations to verify possible adsorption mechanisms. The adsorption characteristics of a material can be expressed in terms of thermodynamic parameters such as ΔG (Gibbs free energy change), which can be calculated by the following equation [17]:

$$\Delta G = -RT \ln K_d \tag{10}$$

where K_d is the thermodynamic equilibrium constant (L·g⁻¹).

According to thermodynamics, the Gibbs free energy is the difference between

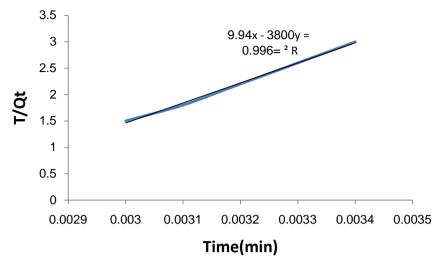


Figure 10. Pseudo second order adsorption kinetics of As(III) onto (SG2P).

Table 4. Pseudo second-order parameters for As(III) adsorption onto SG2P at 25°C.

Adsorbent	Q_{ε} (exp)		pseudo second-order	
	(mg/g)	K_2 min $^{-1}$	Q_e (calculated) (mg/g)	R^2
SG2P	2.3	171.1×10^{-s}	2.1	0.9998

the adsorption enthalpy (ΔH) and adsorption entropy (ΔS) multiplied by the temperature. In this manner, by applying this concept to Equation (9), the thermo chemical parameters ΔH and ΔS can be determined using Van't Hoff's plot (Figure 11), according to Equation (12) [18]:

$$\ln K_d = -\frac{\Delta H}{RT} + \frac{\Delta S}{R} \tag{12}$$

 ΔH° and ΔS° were calculated from the slope and intercept of the linear plot of $\ln K_d$ versus 1/T respectively. The results show that the enthalpy of adsorption ΔH° was -3.7886 kJ·mol⁻¹ and ΔS° was 9.9 J·mol⁻¹·K⁻¹. ΔG° was calculated at different temperatures from the following equation:

$$\Delta G^{\circ} = \Delta H^{\circ} - T \Delta S^{\circ} \tag{13}$$

The obtained thermodynamic values are given in **Table 5**.

The negative ΔG° values indicate that the adsorption is spontaneous at these temperatures. The negative value of ΔH° reflects an exothermic adsorption and indicates that the adsorption is favored at low temperature. In the other hand, the positive value of ΔS° suggests that some structural changes occur on the adsorbent and the randomness at the solid/liquid interface in the adsorption system increases during the adsorption process.

3.9. Antibacterial Activity of SG2P

The results of the minimum inhibitory concentration of micro dilution tray of the examined microorganism are shown in Figure 12 and in Table 6.

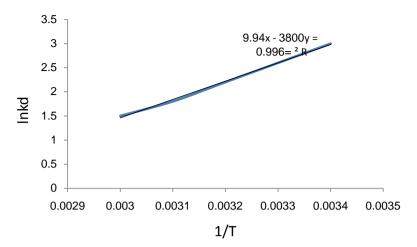


Figure 11. Plot of $\ln K_d$ versus 1/T for As(III) adsorption on (SG2P).



Figure 12. The inhibitory activity of SG2P against *E. coli* and *S. aureus.*

Table 5. The values of the thermodynamic of As(III) adsorption at various temperatures.

Adsorbent	ΔH°	ΔS° (J/mol·K)	ΔG° (KJ/mol)					
	(KJ/mol)		289 K	308 K	318 K	328 K		
SG2P	-3.7886	9.9	-6.6	-6.8	-6.9	-7.0		

Table 6. Demonstration of bacterial growth and inhibition of growth by SG2P in each well.

substance		1	2	3	4	5	6	7	8	9	10	11	12
S. aureus + 25 mg/ml of SG2P	A	-	-	-	-	+	+	+	+	+	+	_	+
S. aureus + 25 mg/ml of SG2P	В	-	-	-	-	+	+	+	+	+	+	-	+
E. coli + 25 mg/ml of SG2P	C	-	-	_	-	+	+	+	+	+	+	-	+
E. coli + 25 mg/ml of SG2P	D	-	-	-	-	+	+	+	+	+	+	_	+
S. aureus	E	_	-	+	+	+	+	+	+	+	+	_	+
S. aureus	F	-	-	+	+	+	+	+	+	+	+	-	+
E. coli	G	-	-	+	+	+	+	+	+	+	+	-	+
E. coli	Н	-	-	+	+	+	+	+	+	+	+	_	+

 $Abbreviation: -, no\ visible\ bacterial\ growth; +, visible\ bacterial\ growth.$

As shown in **Figure 12**, wells in column number 1, 2, 11 and part of 3 and 4 possessed clear broth which indicated absence of bacterial growth (growth inhibition) or inoculation (well 11). On the other hand, wells with turbid broth indicated growth of bacteria due to absence of inhibition of DMSO or SG2P as in positive control of bacterial growth (well 12).

To well number 1 in the first four rows (A, B, C, D), 100 μ l of 25 mg/ml adsorbent (SG2P) dissolved in 50% DMSO was add, thus this well contained 12.5% of adsorbent (SG2P). This was further serially diluted as described in materials and method section. On the other hand, to the first well (number 1) in the remaining four rows (E, F, G, H) 100 μ l of 50% DMSO was added and a serial dilution of DMSO was prepared to detect antibacterial activity of DMSO and to ensure that the bacterial growth inhibition was due to SG2P and not DMSO.

The results shown in **Figure 12**, indicate that the inhibition of both of *E. coli* and *S. aureus* growth occurred in the first four wells containing SG2P concentration of 12.5, 6.25, 3.125 and 1.5625 mg/ml, so the minimum inhibition concentration of SG2P was 1.5625 mg/ml. On the other hand, DMSO inhibited growth of *E. coli* and *S. aureus* in the first two wells (25% and 12.5%) and did not show any antibacterial activity in wells number 3 and 4 in which SG2P showed antibacterial activity. These results indicated that SG2P possessed antibacterial activity against examined bacteria.

4. Conclusion

According to the results obtained in this study, the adsorption capacity of arsenic onto SG2P was efficient. We can be concluded that the removal efficiency of arsenic from aqueous solution has related robustly to operating parameters. The optimum adsorption capacity of arsenic was 50% at pH 5.0, initial concentration 10 μg/L, temperature of 35°C - 40°C and contact time 20 minutes. The adsorption isotherms of arsenic were Freundlich model parameters value 1/n and n, showing that the adsorption of As(III) onto SG2P is promising. While, Lagergren pseudo second order model has been applied to obtain the amount of As(III) adsorbed per unit mass of SG2P, the qe (cal.) was agreement with the experimental value qe (exp.), which explains the exchange between adsorbent and adsorbate. Regarding to antimicrobial activity, SG2P has a low antibacterial activity against both *Staphylococcus aureus*, and *Escherichia coli*. In this paper, we have proposed that it is possible to investigate more studies to explain the adsorption of toxic heavy metals as well as antimicrobial activity onto silica gel compound.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Guan, X.H., Wang, J. and Chusuei, C.C. (2008) Removal of Arsenic from Water

- Using Granular Ferric Hydroxide: Macroscopic and Microscopic Studies. *Journal of Hazardous Materials*, **156**, 178-185. https://doi.org/10.1016/j.jhazmat.2007.12.012
- [2] Ahn, J.S., Chon, C.M., Moon, H.S. and Kim, K.W. (2003) Arsenic Removal Using Steel Manufacturing Byproducts as Permeablereactive Materials in Mine Tailing Containment Systems. *Water Research*, 37, 2478-2488. https://doi.org/10.1016/S0043-1354(02)00637-1
- [3] Korngold, E., Belayev, N. and Aronov, L. (2001) Removal of Arsenic from Drinking Water by Anion Exchange. *Desalination*, 141, 81-84. https://doi.org/10.1016/S0011-9164(01)00391-5
- [4] World Health Organization (WHO) (2006) Guidelines for Drinking Water Quality. Vol. 1, 306.
- [5] Fu, F.L. and Wang, Q. (2011) Removal of Heavy Metal Ions from Wastewater: A Review. *Journal of Environmental Management*, 92, 407-418. https://doi.org/10.1016/j.jenvman.2010.11.011
- [6] Lo, S.L., Jeng, H.T. and Lai, C.H., (1997) Characteristics and Adsorption Properties of an Ironcoated Sand. Water Science & Technology, 35, 63-70. https://doi.org/10.2166/wst.1997.0261
- [7] Shen, Y.S. (1973) Study of Arsenic Removal from Drinking Water. *Journal—American Water Works Association*, 65, 543-548.
 https://doi.org/10.1002/j.1551-8833.1973.tb01892.x
- [8] Worch, E. (2012) Adsorption Technology in Water Treatment-Fundamentals, Processes, and Modeling, Germany. Walter de Gruyter, GmbH & Co. KG, Berlin, 345 p. https://doi.org/10.1515/9783110240238
- [9] Radi, S., Attayibat, A., El-Massaoudi, M., Bacquet, M., Jodeh, SH., Warad, I., Salim S., Al-Showiman, S.S. and Mabkhot, Y.N. (2015) C,N-Bipyrazole Receptor Grafted onto a Porous Silica Surface as a Novel Adsorbent-Based Polymer Hybrid. *Talanta*, 143, 1-6. https://doi.org/10.1016/j.talanta.2015.04.060
- [10] Radi, S., Attayibat, A., Ramdani, A. and Bacquet, M. (2008) Synthesis and Characterization of Novel Silica Gel Supported N-Pyrazole Ligand for Selective Elimination of Hg(II). *European Polymer Journal*, 44, 3163-3168. https://doi.org/10.1016/j.eurpolymj.2008.07.021
- [11] Tiwari, S.K. and Pandey, V.K. (2013) Removal of Arsenic from Drinking Water by Precipitation and Adsorption or Cementation: An Environmental Prospective. *Scienceflora*, **5**, 88-91.
- [12] Attinti, R., Sarkar, D., Barrett, K.R., et al. (2015) Adsorption of Arsenic(V) from Aqueous Solutions by Goethite/Silica Nanocomposite. International Journal of Environmental Science and Technology, 12, 3905-3914. https://doi.org/10.1007/s13762-015-0902-2
- [13] Egger, S., Lehmann, R.P., Height, M.J., Lossener, M.J. and Schuppler, M. (2009) Antimicrobial Properties of a Novel Silver-Silica Nanocomposite Material. *Applied and Environmental Microbiology*, 75, 2973-2976. https://doi.org/10.1128/AEM.01658-08
- [14] Solomon, G. and Anagho, F. (2013) Equilibrium, Kinetic and Thermodynamic Studies of Research Library. *Der Chemica Sinica*, **4**, 58-68.
- [15] Yao, S., Liu, Z. and Shi, Z. (2014) Arsenic Removal from Aqueous Solutions by Adsorption onto Iron Oxide/Activated Carbon Magnetic Composite. *Journal of Environmental Health*, 12, 2-8.
- [16] Hameed, B.H., Mahmoud, D.K. and Ahmad, AL. (2008) Equilibrium Modeling and

- Kinetic Studies on the Adsorption of Basic Dye by a Low-Cost Adsorbent: Coconut (Cocosnucifera) Bunch Waste. *Journal of Hazardous Materials*, **158**, 65-72. https://doi.org/10.1016/j.jhazmat.2008.01.034
- [17] Kubilay, S., Gürkan, R., Savran, A. and Sahan, T., (2007) Removal of Cu(II), Zn(II) and Co(II) Ions from Aqueous Solutions by Adsorption onto Natural Bentonite. *Adsorption*, **13**, 41-51. https://doi.org/10.1007/s10450-007-9003-y
- [18] Umoren, S.A., Etim, U.J. and Israel, A.U. (2013) Adsorption of Methylene Blue from Industrial Effluent Using Poly Vinyl Alcohol. *Journal of Materials and Environmental Science*, **4**, 75-86.