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Sorption of mercury (II) from aqueous solution by Okoume sawdust

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ABSTRACT

In the present study, Okoume sawdust is proposed as a novel sorbent for the removal of mercury (II) from aqueous phase. The operating variables studied were contact time, stirring speed, temperature, solution initial pH and sawdust particle size. The sorption of mercury (II) by Okoume sawdust increased with an increase in contact time, stirring speed and initial solution pH and decreased with an increase in temperature and sorbent particle size. Equilibrium isotherm data were analyzed using the Langmuir and the Freundlich isotherms. The Langmuir model yields a much better fit than the Freundlich model. The maximum sorption capacity of Okoume sawdust was 112.36 mg/g at 20°C. Thermodynamic parameters such as free energy, enthalpy and entropy of sorption have also been calculated. The negative values of the change in free energy and enthalpy indicated that the sorption is spontaneous in nature and exothermic. These results demonstrate that Okoume sawdust is very effective for the removal of mercury(II) from aqueous solutions.

Keywords: Sorption; Valorise of waste; Okoume sawdust; Kinetics; Isotherm; Water treatment; Mercury (II)

1. Introduction

Contamination of the environment from a variety of sources has become an increasingly serious problem in recent years. Heavy metal pollution is an environmental problem of worldwide concern. Heavy metals may come from various industrial sources such as electroplating, metal finishing, textile, storage batteries, lead smelting, mining, plating, and ceramic and glass industries. Lead, chromium and copper are common contaminants of industrial wastewaters. Because they pose serious environmental problems and are dangerous to human health, considerable attention has been paid to methods for their removal from industrial wastewaters [1,2].

The heavy metals are among the most common pollutants found in industrial effluents. Even at low concentrations, these metals can be toxic to organisms, including humans. According to the World Health Organization, the most toxic metals are aluminum, chromium, magnesium, iron, cobalt, nickel, copper, zinc, cadmium, lead and mercury [3].

Mercury, which is a remarkably toxic and non-biodegradable metal, can be generated by several sources, resulting in contamination of atmospheric and aquatic systems. Mercury has very high tendency for binding to proteins and it mainly affects the renal and nervous systems [4]. In humans, the initial symptoms include numbness of the lips and limbs. As the sickness progresses, permanent damage is done to the central nervous system, and the victim experiences visual constriction, loss of motor coordination, and, in the final stages prior to death, loss of memory, speech, hearing and taste. In addition, mercury is easily absorbed through skin, respiratory, and gastrointestinal tissues. The European Union

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considers mercury as a priority and hazardous pollutant and defines a maximum permissible concentration of total mercury as low as 1 μg/l for drinking water and 5 μg/l for wastewater discharge [5,6]. Consequently, removal of mercury ions from water and wastewater is necessary and very important.

Several methods can be applied to remove mercury from aqueous solutions such as precipitation, electrolysis, ion exchange, adsorption, cementation, liquid membranes, and liquid–liquid extraction [7–10]. However, these methods are not widely used due to their high cost and low feasibility for small-scale industries [11]. In contrast, the adsorption technique is one of the preferred methods for removal of heavy metals because of its efficiency and low cost. Conventional adsorbents such as granular or powdered activated carbon are not always popular as they are not economically viable and technically efficient [12]. Non-conventional materials have been tested in a large scale for this purpose such as fly ash [13], lignite [14], tree fern [15], etc. However, the cost of sorbents to be used is the most important restricted factor in view of applicability of sorption process. A number of agricultural waste and by-products have been studied in the literature for their capacity to remove mercury from aqueous solutions [16–20].

In the present study, Okoume sawdust was proposed as a novel material for the sorption of mercury (II) from aqueous phase. It was selected because of its abundance, availability and/or economical relevance. The objectives of this study were to evaluate the removal of mercury (II) ions by Okoume sawdust in batch mode. The effect of operating conditions such as contact time, stirring speed, temperature, solution pH and sawdust particle size on the sorption efficiency of mercury (II) were investigated. The sorption equilibrium data were analyzed using Langmuir and Freundlich models. Additionally, thermodynamic parameters for the sorption of mercury (II) ions were determined.

2. Materials and methods

2.1. Chemicals

Mercury solutions of desired concentration have been prepared by dissolving the appropriate amount of its chloride ($HgCl_2$, Sigma–Aldrich) in distilled water. All chemicals used in this study were of analytical grade.

2.2. Sorbent

In this study, Okoume sawdust was used as a low cost sorbent material for the removal of mercury (II) from aqueous solution. Okoume (*Aucoumea Klaineana*) sawdust is an exotic wood from equatorial West Africa (Gabon). The wood is pink salmon, fibrous, tender often watered with cons-wire bit nervous. It is mainly used for making panels, ply wood, interior carpentry and cigar boxes. The Okoume sawdust was washed with distilled water to remove dust and other materials. The washing process was continued till the wash water contained no color. The washed materials were then completely dried in an oven at 70°C and sieved to eliminate fine particles (>0.5 mm). Finally, the obtained material was stored in a desiccators until use. The principal characteristics of Okoume sawdust were given in Table 1.

Fourier Transform Infrared (FTIR) (SHIMADZU FTIR-408) analysis was applied on the natural Okoume sawdust to determine the surface functional groups, where the spectrum was recorded from 4000 to 400 cm^{-1}. FTIR spectrum for Okoume sawdust is shown in Fig. 1. Fig. 1 shows a strong peak at 3350 cm^{-1} representing the –OH stretching of phenol group of cellulose and lignin, and a peak at 2900 cm^{-1} corresponding to –CH and $-CH_2$ stretching of aliphatic compound. The peaks at 1733 and 1606 m^{-1} were attributed to C=O stretching of aldehyde group and C=C stretching of phenol group, respectively [21,22]. It is clear that the sorbent displays a number of absorption peaks, reflecting the complex nature of the material.

2.3. Kinetic studies

The initial concentration of mercury (II) solution was 100 mg/l for all experiments, except for those carried

Table 1
Principal characteristics of Okoume sawdust

Moisture content (%)	8.316
Apparent specific gravity	0.29
Absolute specific gravity	1.27
Swelling ratio (%)	7.5
PZC	5.64

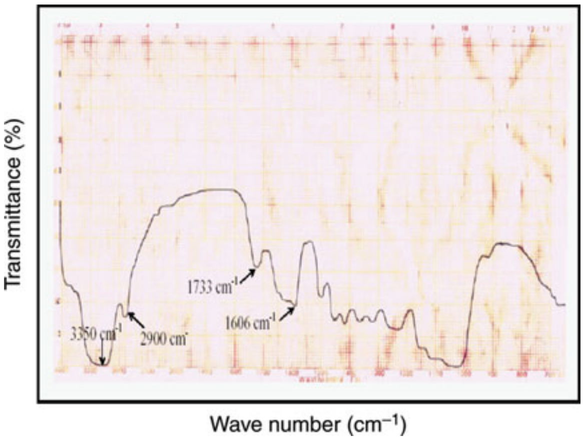

Fig. 1. FTIR spectrum of Okoume sawdust.

out to examine the effect of the initial concentration of mercury. For kinetic studies, the batch method was used because of its simplicity. For mercury removal kinetic experiments, 2 g of Okoume sawdust was contacted with 0.5 l of mercury solution in a beaker agitated vigorously by a stirrer using a water bath maintained at a constant temperature. The stirring speed was kept constant at 300 rpm. At predetermined intervals of time, samples of the mixture were withdrawn at suitable time intervals, filtered through a paper filter and analyzed for the concentration of mercury.

The experiments were performed at the pH that resulted from solving the metal in water (around 5) without further adjustment, except for those conducted to examine the effect of solution pH. Sorption experiments carried out to investigate the effect of pH were conducted by using a volume of 0.5 l of mercury solution of metal concentration 100 mg/l and a sorbent mass of 2 g at 20°C. The solution pH was adjusted using 0.1 N H_2SO_4 or NaOH aqueous solutions. All experiments were repeated at least two times and the mean values were reported.

3. Results and discussion

3.1. Effect of operating conditions

The influence of several operating conditions such as stirring speed, temperature, pH and particle size on the removal of mercury was investigated.

3.1.1. Effect of stirring speed

Fig. 2 shows the amount of mercury sorbed by Okoume sawdust at different stirring speeds ranging from 100 to 800 rpm for a contact time of 90 min. The sorption of mercury by Okoume sawdust was studied by changing the stirring speed while maintaining the initial metal ion concentration (100 mg/l), temperature (20°C), contact time (90 min) and pH 5 constant. The amount of mercury sorption increases from 41.82 to 81.3 mg/g with an increase of stirring speed between 100 and 800 rpm. This can be explained by the fact that increasing stirring speed reduced the film boundary layer surrounding particles, thus increasing the external film transfer coefficient, and hence the fixation of metal. The change in the sorbed amount was insignificant compared to the dissipated energy when the stirring speed increased from 300 to 800 rpm.

3.1.2. Effect of temperature

The temperature dependence of mercury ions was fixed by Okoume sawdust was studied with a constant initial mercury ion concentration of 100 mg/l at 300 rpm at various temperatures (20, 30, 40, and 50°C). Fig. 3 shows the amount of mercury was fixed at equilibrium versus the temperature. It was found that the fixation of metal decreased with the increase in temperature, indicating the exothermic nature of the process. The amount of mercury fixed at equilibrium decreased from 72.2 to 7.7 mg/l with the increasing of temperature from 20 to 50°C, respectively. The removal of heavy metal ions from aqueous solution at various temperatures have been compared with those of other originates sawdust reported in literature. The removal of Cu (II) by cedar sawdust [23] was indicated the exothermic nature of the process. Other investigates were adsorbed the Ni(II), Cu(II), Pd(II) and Cr(II) by meranti sawdust [21] were indicated the endothermic nature of the process.

3.1.3. Effect of solution initial pH

The pH of metal solutions is the major parameter controlling metal sorption processes. This is because hydrogen ion competing with the positively charged metal

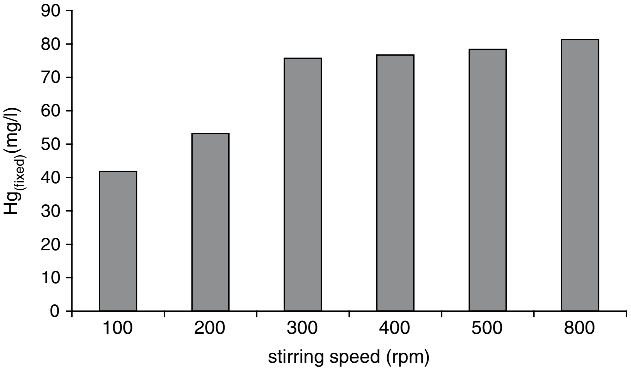

Fig. 2. Effect of stirring speed on the sorption of mercury by Okoume sawdust, conditions: (metal concentration = 100 mg/l; sorbent dosage = 2 g/0.5 l; contact time = 90 min; $T = 20°C$; pH 5, particle size >0.5 mm).

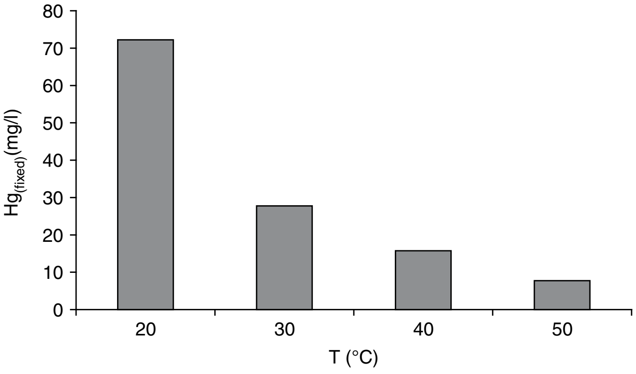

Fig. 3. Effect of temperature on the fixation of mercury by Okoume sawdust (conditions: metal concentration = 100 mg/l; sorbent dosage = 2 g (0.5 l); contact time = 90 min; stirring speed = 300 rpm; pH 5, $d > 0.5$ mm).

ions on the active sites of the sorbent. From Fig. 4, it was observed that the fixation was strongly pH-dependent. The maximum fixation of metal takes place at pH 8.

The variation in the removal of mercury by Okoume sawdust with respect to pH can be elucidated by considering the surface charge of the sorbent materials and the speciation of mercury (II). According to the Hg(II) speciation diagram [24] at low pH values (3 and 4), the lower fixation of mercury at acidic pH is due to the presence of excess H^+ ions competing with metal ions for the sorption sites. The PZC of Okoume sawdust is 5.64. The surface charge of Okoume sawdust is positive at pH <5.64, is neutral at pH 5.64, and is negative at pH >5.64. At high pH values (6–10), greater than 5.64, Okoume sawdust becomes negatively charged and the Hg(II) species are still present mainly as $Hg(OH)^+$ and $Hg(OH)_2$ at pH 6 and 7 and as $Hg(OH)_2$ at pH 8–10. The effect of pH on the sorption kinetics is attributed to electrostatic attraction existing between the Okoume sawdust of surface and the Hg^{2+} ions in solution.

3.1.4. Effect of the particles size

In order to study the effect of this parameter on the metal fixation by Okoume sawdust, the diameter of sobent was varied from 0.08 mm at 1.25 mm. The specific area of sorbent was imported for the rate of sorption metal ions. The particles size of wood flour plays a very important role in the fixation of metal ions, it reveals that the smaller particle size can increase the exchange surface along the porous material, facilitates access to the active sites of the biosorbent responsible for binding [25].

From Fig. 5, it was observed that the mercury removal varied with varying diameter of sawdust of Okoume. It was noticed that an decrease in diameter of sorbent to an increase in the fixation capacity of mercury by Okoume sawdust this is because a large specific area of sorbent.

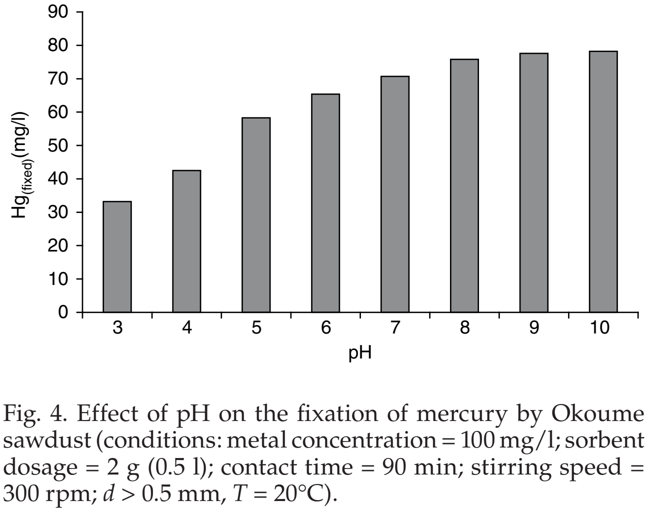

Fig. 4. Effect of pH on the fixation of mercury by Okoume sawdust (conditions: metal concentration = 100 mg/l; sorbent dosage = 2 g (0.5 l); contact time = 90 min; stirring speed = 300 rpm; $d > 0.5$ mm, $T = 20$°C).

3.2. Equilibrium isotherms

Equilibrium isotherms were determined in (Fig. 6) by contacting a fixed mass of Okoume sawdust (2 g) with 0.5 l of mercury(II) solutions in beakers. A range of mercury(II) concentrations (50–600 mg/l) was tested. A series of such beakers was then stirred using mechanical stirrers at a constant speed of 300 rpm in a water bath with temperatures 20, 30, 40, and 50°C, respectively. After agitating the beakers for 3 h, the reaction mixtures were filtered through filter paper then the filtrates were analyzed. All experiments were repeated and the mean values have been reported. The experiments were performed at pH 5, which resulted from dissolution of mercury chloride in distilled water without further adjustment. The amount of sorption (fixation) at equilibrium, q_e (mg/g), was calculated by equation [26]:

$$q_e = \frac{(C_0 - C_e) \times V}{m} \tag{1}$$

where q_e is the adsorption capacity (mg/g), C_0 and C_e (mg/l) are the initial and equilibrium liquid phase

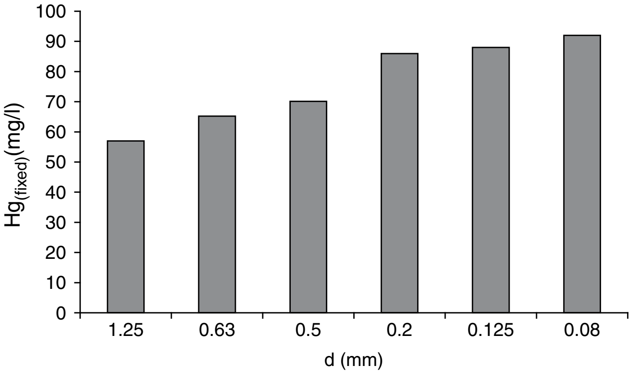

Fig. 5. Effect of diameter of sawdust of Okoume on fixation of mercury (conditions: metal concentration = 100 mg/l; sorbent dosage = 2 g (0.5 l); contact time = 90 min; stirring speed = 300 rpm; $T = 20$°C).

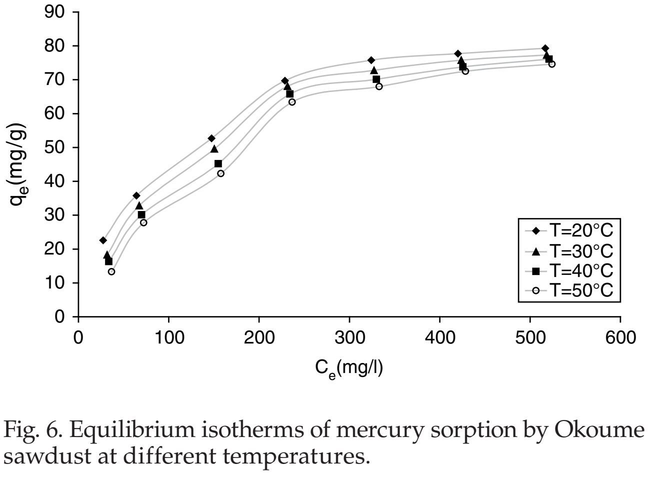

Fig. 6. Equilibrium isotherms of mercury sorption by Okoume sawdust at different temperatures.

concentrations of metal, respectively. V (l) is the volume of the solution and m (g) is the mass of the dry sorbent used.

The sorption isotherm of mercury was determined for sorbent. The removal of mercury has been studied at 20, 30, 40, and 50°C to determine the sorption isotherm and thermodynamic parameters, which is presented in Fig. 6. Isotherm data obtained with a range of initial Hg (II) concentration showed an increase in the amount of Hg (II) sorbed when the initial metal concentration was raised from 50 to 600 mg/l. Additionally, the amount of metal sorbed decreased following an increase in temperature from 20 to 50°C. The shape of the curves clearly indicated that the isotherms for all temperatures belong to L type according to the classification of equilibrium isotherm in solution by Giles et al. [27]. To quantify the adsorption capacity of Okoume sawdust for the removal of Hg (II) ions from aqueous solution, the Langmuir and Freundlich isotherm models were used.

3.2.1. Langmuir model

This model assumes that the adsorptions occur at specific homogeneous sites on the adsorbent and is used successfully in many monolayer adsorption processes. The data of the equilibrium studies for adsorption of Hg (II) ions on to Okoume sawdust at 20, 30, 40, and 50°C may follow the following form of Langmuir model:

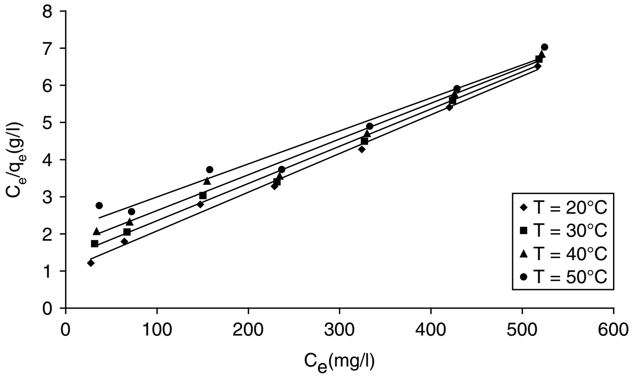

Fig. 7. Sorption isotherms for mercury by Okoume sawdust at 20, 30, 40, and 50°C (Langmuir isotherm).

$$\frac{C_e}{q_e} = \frac{1}{b \cdot q_m} + \frac{1}{q_m} \times C_e \tag{2}$$

where C_e (mg/l) and q_e (mg/g) are the liquid phase concentration and solid phase concentration of sorbate at equilibrium, respectively; b (l/mg) is the Langmuir isotherm constant; q_m (mg/g) is the maximum sorption capacity of Langmuir. Hence, a plot of C_e/q_e vs. C_e (Fig. 7) should be a straight line with a slope $(1/q_m)$ and an intercept as $(1/q_m b)$.

The values of constants q_m and b were calculated and reported in Table 2. A further analysis of the Langmuir equation can be made on the basis of a dimensionless equilibrium parameter, R_L [28,29], also known as the separation factor, given by:

$$R_L = \frac{1}{1 + b \times C_0} \tag{3}$$

where b is the Langmuir constant and C_0 is the initial concentration of the sorbate in solution.

The value of R_L lies between 0 and 1 for favorable adsorption, while $R_L > 1$ represents unfavorable adsorption, and $R_L = 1$ represents linear adsorption while the adsorption process is irreversible if $R_L = 0$. The calculated R_L values versus initial mercury concentration at four different temperatures were represented in Fig. 8. From this figure, it was observed that sorption was found to be more favorable at higher concentrations. Also the value of R_L in the range of 0–1 at all initial metal concentrations and the four studied temperatures confirms the favorable uptake of mercury process.

3.2.2. Freundlich model

The Freundlich model can be applied for non-ideal sorption on heterogeneous surfaces and multilayer sorption. According to this model:

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \tag{4}$$

Table 2
Parameters of the Langmuir and Freundlich isotherm models

T (°C)	Langmuir			Freundlich		
	q_m (mg/g)	b (l/mg)	r	K_F ($\text{mg}^{1-(1/n)}\ \text{l}^{1/n}$/g)	n	r
20	112.36	0.475	0.984	5.55	2.28	0.986
30	104.16	0.601	0.993	3.40	1.92	0.977
40	100.00	0.734	0.996	2.49	1.79	0.981
50	96.15	0.965	0.997	1.55	1.56	0.976

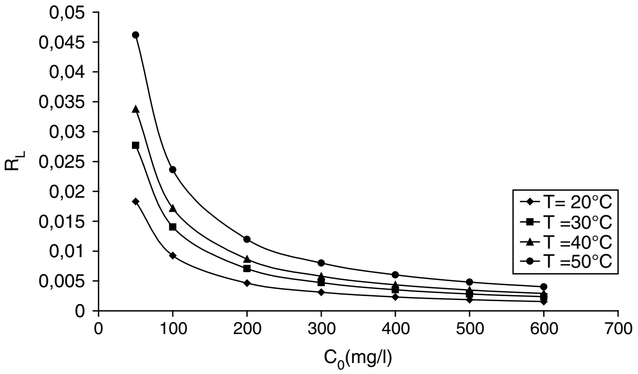

Fig. 8. Separation factor for the uptake of mercury by Okoume sawdust at 20, 30, 40, and 50°C.

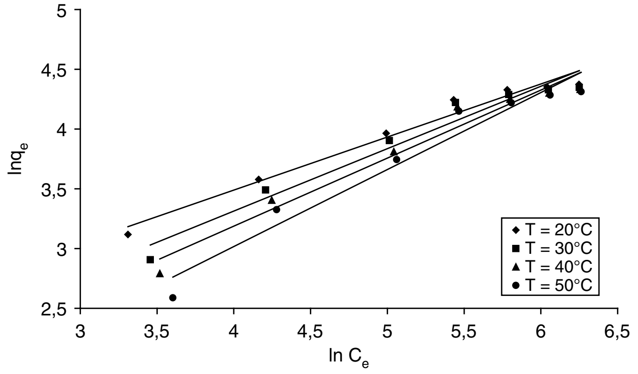

Fig. 9. Sorption isotherms for mercury by Okoume sawdust at 20, 30, 40, and 50°C (Freundlich isotherm).

where q_e is the amount sorbed (mg/g), C_e is the equilibrium concentration of the sorbate (mg/l), and K_F ($mg^{1-1/n}$ $L^{1/n}$/g) and n are Freundlich constants related to the sorption capacity and sorption intensity, respectively. If the Freundlich equation fits the sorption isotherm, K_F and n can be obtained from the plot of ln q_e vs. ln C_e (Fig. 9) .Freundlich constants K_F and n are calculated from plots, and their values at four different temperatures, 20, 30, 40, and 50°C, are given in Table 2.

The Freundlich type adsorption isotherm is an indication of surface heterogeneity of the adsorbent while Langmuir type isotherm hints towards surface homogeneity of the adsorbent. This leads to the conclusion that the surface of Okoume sawdust is made up of small heterogeneous adsorption patches which are very much similar to each other in respect of adsorption phenomenon.

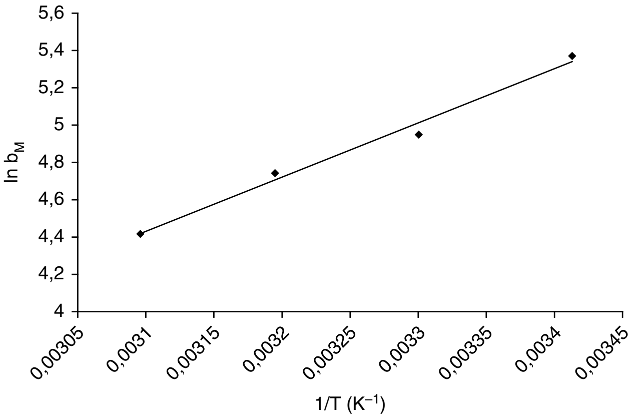

Fig. 10. Plot of ln b_M versus $1/T$ for the determination of thermodynamic parameters (Okoume sawdust).

Table 3
Thermodynamic parameters for the sorption of mercury by Okoume sawdust

T (°C)	20	30	40	50
ΔG° (kJ/mol)	–13.08	–12.46	–12.34	–11.86
ΔH° (kJ/mol)	–24.17			
ΔS° (J/mol/K)	–37.8	–38.6	–37.7	–38.1

3.3. Thermodynamic parameters

In engineering practice, entropy and energy factors should be considered in order to determine what processes will occur spontaneously. Thermodynamic parameters for the sorption systems, i.e., free energy ($\Delta G°$), enthalpy ($\Delta H°$), and entropy ($\Delta S°$), changes were calculated by using the following equations [30]:

$$\ln b_M = \frac{\Delta S°}{R_g} - \frac{\Delta H°}{RT} \tag{5}$$

$$\Delta G° = \Delta H° - T\Delta S° \tag{6}$$

where T (K) is the absolute temperature, R_g (kJ/mol/K) is the gas constant, and b_M (l/mol) is the Langmuir equilibrium constant. It is known that (ΔG°) is the function of change in enthalpy of sorption ($\Delta H°$) as well as change in standard entropy ($\Delta S°$).

By using Eq. (5) the values of $\Delta H°$ and $\Delta S°$ of mercury sorption by the studied sorbent were determined from the slope and the intercept of the linear plot of ln b_M vs. $1/T$ (Fig. 10). The Gibbs free energy change ($\Delta G°$) was obtained according to Eq. (6).

The thermodynamic parameters obtained for the sorption of mercury by Okoume are given in of Table 3. The negative values of $\Delta G°$ demonstrate spontaneous and favorable sorption process. The negative values of enthalpy change (–24.17 kJ/mol**)** is due to the exothermic nature of sorption. The low value of $\Delta S°$ may imply that no remarkable change in entropy occurred during the sorption of mercury by the material. In addition, the negative value of $\Delta S°$ reflects the decreased randomness at the solid–solution interface during sorption of the sorbent toward mercury. Normally, adsorption of gases leads to a decrease in entropy due to orderly arrangement of the gas molecules on a solid surface. However, the same may not be true for the complicated system of sorption from solution onto Okoume sawdust. Some researchers have indicated that the positive values of $\Delta S°$ suggested some structural changes in metal and sorbent [31,32].

4. Conclusions

The results obtained in this study demonstrated the potential use of Okoume sawdust for the sorption (fixation) of Hg (II) ions from aqueous solutions. The kinetic studies indicated that equilibrium in the sorption of Hg (II) ions on Okoume sawdust was reached in 90 min of contact between the sawdust and the solution. It was found that the amount of Hg(II) metal ions sorbed into the Okoume sawdust increased with an increase in stirring speed and solution initial PH. In this study, the effect of temperature and the particles size was revealed that fixation (sorption) of metal ions Hg (II) by Okoume sawdust increased with a decrease of temperature from 50 to 20°C and the diameter from 1.25 to 0.08 mm. The Langmuir model yielded a much better fit than the Freundlich model. The maximum sorption capacity was determined to be 112.36, 104.16, 100.00, and 96.15 mg/g at 20, 30, 40, and 50°C, respectively. The R_L values showed that Okoume sawdust was favorable for the sorption of mercury. The negative values of free energy change negative value of enthalpy change and entropy indicated that the Hg (II) sorption process is spontaneous, exothermic and reflect the decreased randomness at the solid–solution interface during sorption of the sorbent toward mercury respectively.

Symbols

b — Langmuir constant related to the free energy of sorption (l mg^{-1});
b_M — Langmuir constant related to the free energy of sorption (l mol^{-1});
C_e — the equilibrium concentration of the solute in the bulk solution (mg l^{-1});
C_0 — the initial concentration of the solute in the bulk solution (mg l^{-1});
C_r — the residual concentration of the solute in the bulk solution (mg l^{-1});
C_u — coefficient of uniformity ($C_u = D_{60}/D_{10}$);
D_m — the mean diameter;
D_e — the effective diameter;
K_F — Freundlich constant indicative of the relative sorption capacity of the sorbent (mg $^{1-(1/n)}$l$^{1/n}$/g);
m — sorbent mass (g);
n — Freundlich constant indicative of the intensity of the sorption;
q — the amount of solute sorbed at any time t (mg/g) ;
q_e — the amount of solute sorbed per unit weight of sorbent at equilibrium (mg/g);
q_m — the maximum sorption capacity (mg/g);
R — coefficient of correlation;
R_g — universal gas constant (J/mol/K);
R_L — dimensionless separation factor of Hall;
t — time (min);
T — temperature (K);
V — solution volume (l);
$\Delta G°$ — Gibb's free energy change (kJ/mol);
$\Delta H°$ — enthalpy change (kJ/mol);
$\Delta S°$ — entropy change (J/mol/K);
PZC — point of zero charge

Greek letters

ε — Porosity of sorbent;
ρ — Density of sorbent, g/cm

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