

Optimization of Phenol Adsorption by the Fine Fraction of Mouyondzi and Missafou Clay

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Abstract Industrial development significantly meets the needs of our society. This results in significant releases of pollutants into the environment. Petrochemical and oil refineries are among the industries handling products with the highest polluting capacity. Oil remains the most widely used energy source, particularly for transportation. To meet this demand, crude oil is processed and transformed, the goal being to extract as many high-value products as possible. However, refining processes pose a health risk. Oil refineries produce significant quantities of wastewater during their operations. This wastewater typically contains various types of pollutants, including heavy metals, organic compounds, and other toxic substances. However, oil refineries often have on-site water treatment plants to mitigate the environmental impact of their effluent. These plants use various physical, chemical, and biological processes to remove pollutants from wastewater before it is discharged into waterways or reused elsewhere in the refinery. However, clays have also shown adsorption capacity, degreasing and decolorizing water polluted by industrial discharges. This study is based on this perspective. It reports the adsorption of phenol by local clay materials. Indeed, if it is clearly accepted that Congo has oil; it is also known that the refining process of the latter releases chemicals, in this case phenol. The latter is toxic in the ambient environment and can contaminate surface water, soils and groundwater. The main objective of this study is to enhance the adsorbent properties of local clay materials in the retention of phenol likely to be present in soils and surface waters. To do this, we chose talc from Missafou and kaolinite from Mouyondzi, two localities in the Republic of Congo. The fine fraction is extracted by the method described by Thierry Holtzapffel. Kinetic data showed that phenol was bound after 20 minutes of contact for Mouyondzi kaolinite and 30 minutes for Missafou talc. Batch adsorption tests showed that the adsorption capacity of the clays used was quite high for phenol, and that the Mouyondzi fine fraction adsorbed better than that of Missafou (1.26 mg/g and 1.07 mg/g, respectively), while the adsorption isotherms were more satisfactorily described by the Langmuir model. Both clays were suitable for use as adsorbents for pollution control.

Keywords: adsorption, phenol, clay, Mouyondzi, Missafou

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1. Introduction

Phenols are among the products used in industrial manufacturing activities such as the manufacture of paper, plastics, dyes, pharmaceuticals, and agrochemicals. They are present in effluents from refineries, coke ovens, petrochemical plants, etc. Known to be toxic to humans and the environment, their taste and odor pose significant

problems even at very low concentrations and are currently attracting particular international attention [1]. To protect human health from the potential toxic effects caused by exposure to phenol, it is therefore essential to eliminate it from various industrial effluents or reduce their quantity below the permissible thresholds defined by standards before releasing them into the natural environment. To this end, there are several methods for treating phenol in industrial effluents. Physicochemical treatment methods have proven to be expensive and have

inherent drawbacks due to the formation of toxic by-products such as chlorinated phenols. For this reason, research has focused on treatment processes using natural, easily exploitable and inexpensive materials [2]. Various studies have shown that adsorption on clays is an effective treatment process for removing a very wide variety of toxic compounds from the environment. Kaolinite, for example, is suitable for the sorption of fluoride ions from water. Radioactive alkali metals are more efficiently absorbed by clay minerals (mica), while chlorite is suitable for divalent radionuclides. A mixture of exfoliated vermiculite with treated calcium bentonite and peat can be used as a deodorizing sorbent. Calcium bentonite is used as a nutrient sorbent from water in dams and other reservoirs [3,4]. In this work, we were interested in evaluating the adsorption capacity of Missafou talc and Mouyondzi kaolinite (two localities in the south of the Republic of Congo) with respect to phenol. Although the 1:1 clay species have a low cation exchange capacity, their abundance in the region requires numerous applications [5,6,7,8,9]. What is then the adsorption capacity of these clays on the elimination of phenol?

2. Materials and Methods

2.1. Description and Location of Sampling Sites

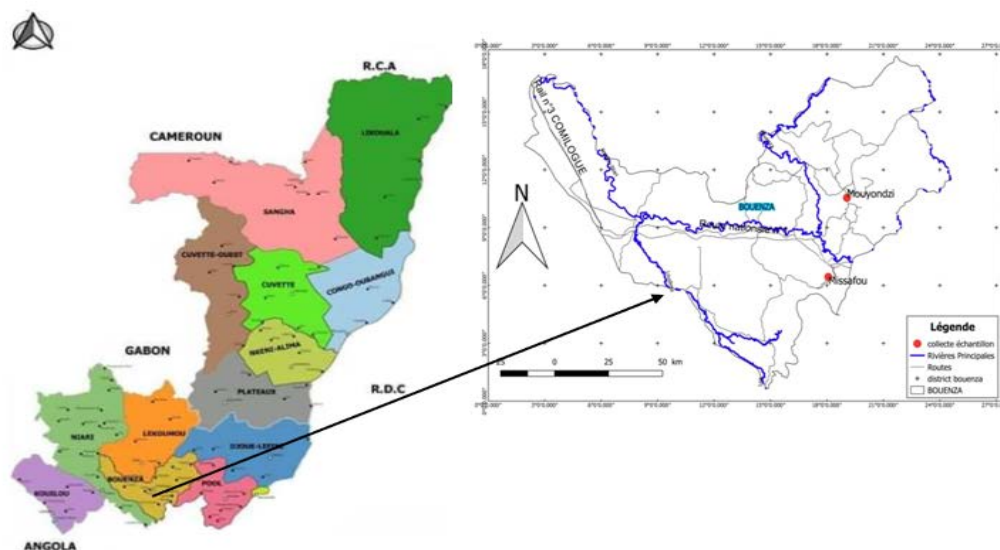


Figure 1. Location of the sampling site [11]

- Moutou et al reported that the dominant clay species in Mouyondzi is kaolinite, the main impurity is quartz [5]. Numerical data deduced from the N_2 adsorption/desorption isotherm reveal that Mouyondzi clay has a fairly large pore volume. Indeed, it reaches a value of 110 110 cm^3/g [12].
- Kouhounina et al prove that Missafou talc and Mouyondzi kaolinite show an affinity for glycine. The retention capacity is 9.12 mg/g for Missafou while that of Mouyondzi is 10.77 mg/g. The adsorption isotherms were modeled by the Langmuir and Freundlich models. The models

The sampling sites are located in the Bouenza Department. The Bouenza Department is subdivided into ten (10) districts: Boko-Songho, Kayes, Kingoue, Loudima, Mabombo, Madingou, Mfouati, Mouyondzi, Tsiaki, and Yamba. Its capital is Madingou. One of the clay materials used in this study comes from the Mouyondzi district, specifically from the village of Nzaou. The other comes from the Mfouati district, specifically from the village of Missafou. The clay from the Nzaou deposit is widely consumed by pregnant women and used by traditional healers to treat various diseases. The Bouenza Department is located midway between Brazzaville and Pointe-Noire. Crossed by the Congo-Ocean Railway for nearly 150 km, it is bordered to the north by the Lekoumou department, to the south by the Democratic Republic of Congo, to the east by the Pool department, and to the west by the Niari department. The Niari Valley, in which the Bouenza department is located, has soils whose parent rock consists of schist-limestones represented by pink and gray dolomites, clayey limestones, built-up limestones and crystalline limestones, pebbly sands, and sandy limestones [10]

The geographical coordinates shown in Table 1 allowed us to locate the sampling sites.

Table 1. Geographic coordinates

Sample	Latitude	Longitude	Altitude
Missafou	9518750	0373551	235 m
Mouyondzi	9554532	0382155	410 m

showed that adsorption is favorable with the Langmuir model [12].

2.2. Products Used

In our study, hydrochloric acid was used to destroy carbonates to allow the suspension of clay particles. Oxalic acid was used to remove iron oxides, and hydrogen peroxide was used to destroy organic matter. Like carbonates, these substances also agglomerate clay particles, preventing their dispersion. Adsorption tests were conducted with phenol, a commercially available product from PROLABO. The chemical structure, molar

mass, and physicochemical characteristics of the selected pollutant are summarized in Table 2.

Table 2. Physicochemical characteristics of phenol.

Chemical Name	Phénol
Molecular Formula	C ₆ H ₆ O
Molar Mass (g)	94.11
Boiling Point	181.75°C
Melting Point	40.8°C
Solubility in Water	8.2 g /100ml
Flash Point	82°C
Relative Density	1.07g /cm ³ à 25°C

2.3. Purification and Extraction of the Fine Fraction (2 µM)

The method used for this operation is that described by Thierry Holtzapffel [13]. The clays were separated through a series of successive decantations. After disintegration, the resulting suspension was first treated to remove the cements accompanying the clays in the soil. Carbonates were removed using dilute hydrochloric acid (N/5). The pH was monitored throughout the reaction using universal pH paper. When the pH turned pink, indicating that the carbonates were dissolved, the reaction was stopped, and the suspensions were then diluted in distilled water. The organic matter was destroyed using hydrogen peroxide (20 volumes) mL per mL in a water bath at a temperature below 60°C. This treatment is stopped when the release of bubbles has decreased and the color has lightened. The elimination of iron oxides was carried out by 20% oxalic acid, hot (<60°C) in a water bath. Once the color in the Erlenmeyer flask has become greenish, the treatment is stopped, the suspensions are then washed by centrifugation until the disappearance of this greenish color which reflects the presence of ferrous ions. The extraction of the fine fraction was done by sedimentometry.

Table 3. Sedimentation time required for sampling

Sampled particle size fraction	Sampling depth	
	1 cm	2cm
2µm	48mim	1h35mim

2.4. Verification of Beer-Lambert's Law

To perform the calibration, we prepared various phenol solutions of decreasing concentration by dilution from a stock solution of phenol with a concentration of 753 mg/L. These were then analyzed using ZUZI UV-visible spectrophotometry. This established the calibration line representing the optical density at the maximum of the adsorption band as a function of concentration.

2.5. Adsorption Kinetics

The aim of this study was to determine the time required for optimal adsorption. The phenol concentration was 10-3 mol/L, and the adsorbent masses were identical (0.5 g). The suspensions were stirred for different times and then centrifuged for 30 minutes at 6000 rpm. The

supernatant was then immediately measured using a UV-visible spectrophotometer at a wavelength of 270 nm. The amount adsorbed is determined by the following relationship:

$$Q = \frac{(C_0 - C_t) \cdot V}{m}$$

Q: the fixed quantity of pollutant in mg per gram of adsorbent,

C₀ and C_t: are respectively the initial and instantaneous concentrations of the pollutant (mg/l),

V: the volume of the solution (L),

m: the mass of the adsorbent used (g).

2.6. Adsorption Tests

The batch method is used. Based on the equilibrium time obtained, adsorption is performed by taking 0.5 g of adsorbent and 10 mL of the phenol solution at different concentrations (94, 47, 24, 12, and 6 mg/L), and stirring for thirty (30) minutes. After centrifugation, the supernatants are then measured by UV-visible spectroscopy at a wavelength of 270 nm.

2.7. Evaluation of Adsorbent Power

Two mathematical models are used for this study: the Langmuir model and the Freundlich model.

For the Freundlich model, the equation obtained is that of a straight line with slope 1/n and y-intercept Ln K_f, from which the Freundlich constants K_f and n are derived.

For the Langmuir model, this is the equation of a straight line with slope $\frac{1}{Q_{\max} \cdot K_L}$ and y-intercept $\frac{1}{Q_{\max}}$, which allows the determination of the two equilibrium parameters Q_{max} and K_L.

The shape of the Langmuir isotherm is indicated by a dimensionless term R_L called the separation factor or equilibrium parameter, defined by:

$$R_L = \frac{1}{1 + K_L \times C_0}$$

- K_L: the thermodynamic constant of the adsorption equilibrium characteristic of the adsorbent, dependent on temperature and experimental conditions (in l.mg-1)
- C₀, the concentration of the phenol solution [14].

For both adsorbents used in this study, this factor ranges between 0 and 1, indicating favorable adsorption.

3. Results and Discussion

3.1. Phenol Calibration Curve

Figure 2 shows the calibration curve plotted by plotting absorbance versus concentration to verify the Beer-Lambert law.

This curve is linear with a correlation coefficient (R=0.9720) close to 1. The chosen concentration range satisfies the Beer-Lambert law.

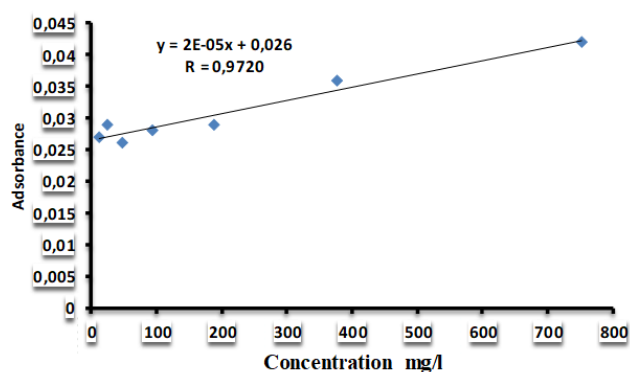


Figure 2. Phenol calibration curve

3.2. Adsorption of phenol by Missafou and Mouyondzi clay

1) Adsorption kinetics and isotherm

a. Adsorption kinetics by Missafou and Mouyondzi

Figure 3 and Figure 4 represent the adsorbed quantities of phenol as a function of contact time by the fine fraction of Missafou and Mouyondzi, respectively.

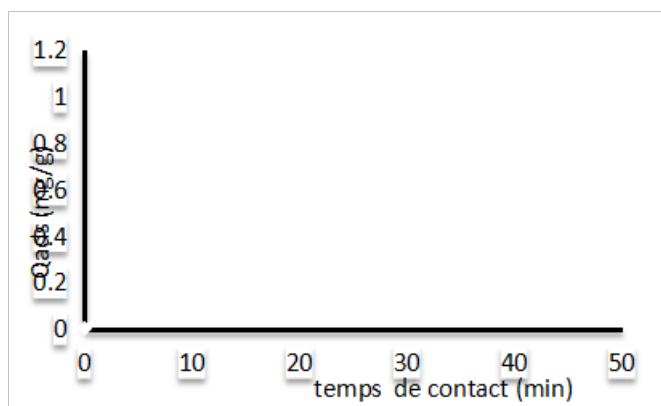


Figure 3. Adsorption kinetics of Phenol on MISA

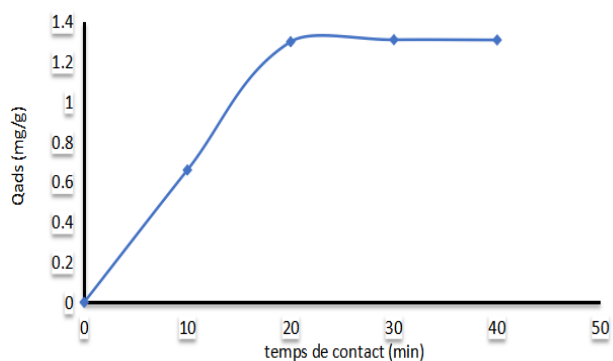


Figure 4. Adsorption kinetics of Phenol on MOU

The evolution of phenol removal curves by clays can be broken down into two phases: a very rapid first, followed by a medium-speed second, to reach the saturation plateau. This phenomenon can be explained in the first stage by the existence of easily accessible adsorption sites, followed by diffusion towards less accessible adsorption sites due to the existence of repulsive forces between the phenol molecules adsorbed by the materials and those in solution before reaching adsorption equilibrium where all sites

become occupied. For the MOUYONDZI clay, fixation is rapid ($t = 20$ minutes) compared to that of Missafou ($t = 30$ minutes). For the rest of our experiments, the solid-solution contact time will be fixed at these different times.

b. Adsorption isotherm by Missafou and Mouyondzi

Adsorption isotherms are very useful for understanding the adsorption mechanism. These adsorption isotherms are represented by plotting the amount adsorbed at equilibrium as a function of concentration.

Figure 5 and Figure 6 show the adsorption isotherms of phenol by the Mouyondzi and Missafou clays.

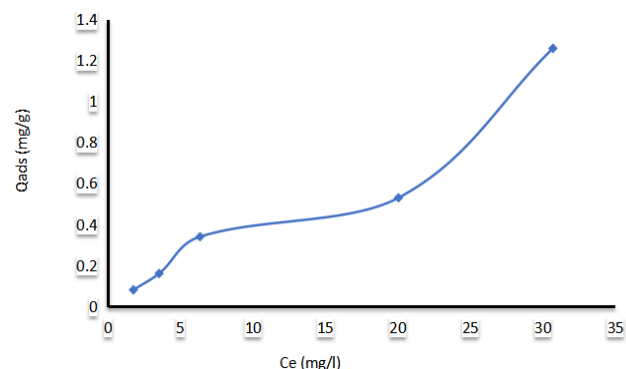


Figure 5. Adsorption isotherm on Mou

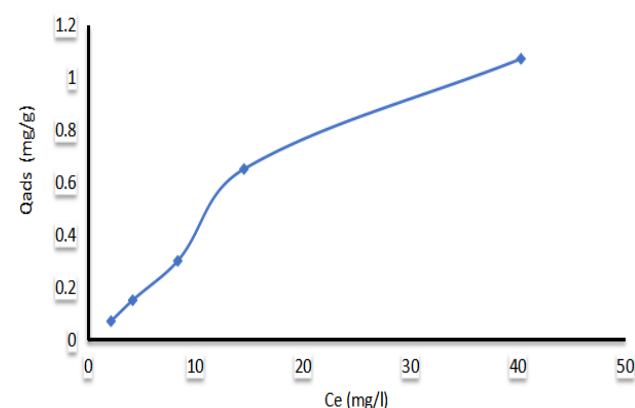


Figure 6. Adsorption isotherm on MISA

Analysis of these curves shows that the adsorbed quantities increase with increasing equilibrium concentrations of phenol (C_e). This behavior can be explained by the presence of a large number of phenol ions in solution, which would imply massive adsorption within the materials, thus leading to an increase in the adsorption quantity of MISA and MOU.

The adsorption isotherm on MOU is of type S, which would reflect, on the one hand, vertical adsorption as is the case for molecules with a single functional group and, on the other hand, when the molecules are in strong adsorption competition with the solvent [15]. It is of type L on MISA, in this case the adsorption of the solvent is weak and that of the solute on the solid is in a monolayer.

We also observe an inflection of the curves without reaching a plateau. Clays therefore tend, under our conditions, to retain relatively larger quantities of phenol. The highest adsorbed quantities are 1.26 mg/g for MOU and 1.07 mg/g for MISA. These results are in agreement with the presence of a higher specific surface area and

percentage of fine particles for mouyondzi [5,12]. The same observation was made in the work carried out by Henni-Chebra Fatma El Batoul on the elimination of industrial pollutants by lamellar double hydroxides (LDH) [16].

3.3. Adsorption Model

To evaluate the adsorption results, the linearized forms of the phenol isotherms on MOU and MISA are given in the following figures:

1) Langmuir model of Mou and MISA

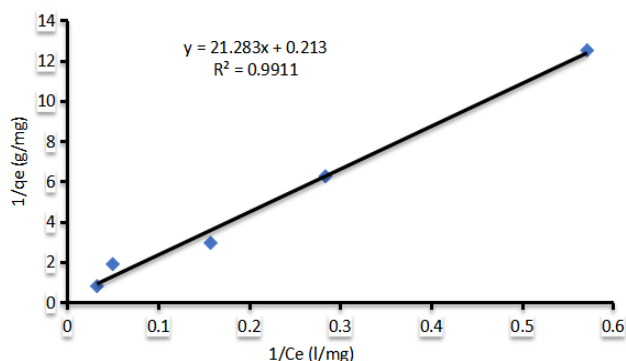


Figure 7. Application of the Langmuir model for the adsorption isotherm of phenol by MOU

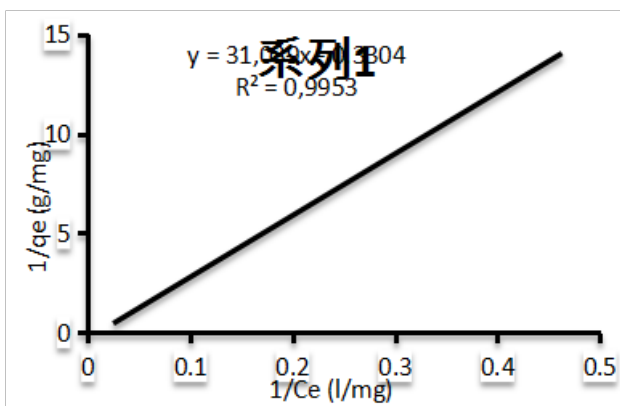


Figure 8. Application of the Langmuir model for the adsorption isotherm of phenol by MISA

2) Freundlich model for MOU and MISA

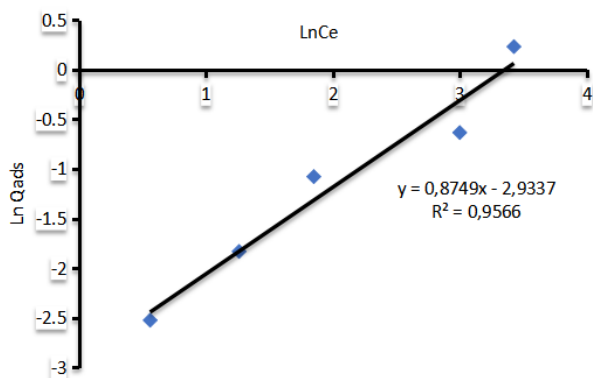


Figure 9. Application of the Freundlich model for the adsorption isotherm of phenol by MOU

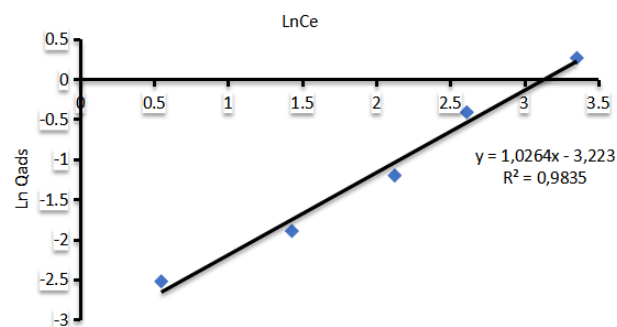


Figure 10. Application of the Freundlich model for the adsorption isotherm of phenol by MISA

Table 4. Different Langmuir and Freundlich coefficients obtained from experimental results, for phenol on the fine fraction of Mouyondzi and Missafou

Clay materials	Langmuir			Freundlich		
	$Q_{\max}$ (mg/g)	K_L (L/mg)	R	$1/n$	K_F	R
MISA	2.85	0.015	0.997	1.01	0.7	0.98
MOU	4.69	0.010	0.995	0.8749	0.05	0.97

The $1/n$ parameter is 1.01 for MISA and 0.8749 for MOU. The determined values of n are 0.99 and 1.14 for MISA and MOU, respectively. It has been reported in the literature that adsorption is favorable for values of $n > 1$, which allows us to say that the adsorption of phenol is favorable with Mouyondzi kaolinite [17]. This confirms its maximum adsorption capacity $Q_{\max}$ which is 4.69 mg/g. This value is higher than that of MISA ($Q_{\max} = 2.85$ mg/g); a consequence of the results of their specific surface area. Adsorption therefore depends on both the nature of the adsorbate and that of the adsorbent material.

The Freundlich distribution coefficient K_F is relative to the total sorption capacity of the solid; it is 0.7 for MISA and 0.05 for MOU. K_L which is the thermodynamic constant of the adsorption equilibrium characteristic of adsorbent is 0.015 for MISA and 0.010 for MOU and R which is the correlation coefficient are all close to unity with both models. This induces an affinity between phenol and the materials used.

4. Conclusion

This work was devoted to evaluating the adsorption capacity of two clay soils for phenol. To achieve this objective, adsorption kinetics and isotherms were performed using the batch method. Two models were used to model the adsorption isotherms, namely the Langmuir and Freundlich models. The results obtained show that the adsorption capacity of Mouyondzi clay is higher than that of Missafou clay, with maximum adsorption amounts of 4.69 mg/g and 2.85 mg/g for Mouyondzi and Missafou, respectively. Based on the correlation coefficient R , related to the linearity of the adsorption isotherm lines, the adsorption process follows both models. The high values of the Freundlich constants K_F and $1/n$ allow us to conclude that the adsorption isotherms of phenol on the

clays of Mouyondzi and Missafou are better represented by Langmuir. It has been demonstrated through this study that kaolinite, which is a 1:1 clay species, can also eliminate pollutants. The use of clays in the context of decontamination will not only contribute to the preservation of the environment but also reduce the cost compared to other decontamination methods used. To increase their adsorption capacity, activation will be considered.

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