Removal of Phenolphthalein from Laboratory Wastewater Using Natural Clay and Activated Carbon

Abderezak Guemache*

Laboratory of Water, Environment and Renewable Energies (LWER), Faculty of Technology, University of M'sila, Pole, Road Bordj Bou Arreridj, M'sila 28000, Algeria

* ***Corresponding author:***

email: abderezak.guemache@univ-msila.dz

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Abstract: *Dyes from laboratory wastewater are considered a potential source of water contamination. In this study, natural clay and activated carbon were used to remove a colored indicator (phenolphthalein). Both adsorbents were analyzed by X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR) and the colored indicator was characterized by UV-vis spectrophotometer. The effects of various parameters, such as initial phenolphthalein concentration, contact time, temperature, pH, and decolorization, were studied. Dye removal increased with decreasing initial phenolphthalein concentration and solution contact time. The percentage of phenolphthalein removal increased accordingly, reaching 99% for activated carbon and 98% for natural clay. Langmuir and the Freundlich adsorption models were used to describe the adsorption equilibrium. The data very well fitted with these models. The monolayer adsorption capacities were equal to 31 mg g^{-1} at pH 8.0 and temperature 27 °C. The adsorption measurements show that the adsorption process is rapid and physical in nature. The results explain that the adsorption process isexothermic and spontaneous physisorption.*

Keywords: *adsorption; phenolphthalein; kinetic model; natural clay; activated carbon*

■ INTRODUCTION

Water is a fundamental element in the maintenance of life and it is indispensable to existence. This resource is used in many sectors, including industry, but once used, water is often discharged into the natural environment, which causes aquatic pollution [1]. Pollution is a serious problem, affecting virtually all ecosystems, including the aquatic environment. The main water pollutants are organic pollutants (industrial and domestic). The latter are persistent and toxic, for which several treatment methods have been developed to eliminate them [2].

The management and treatment of laboratory wastewater contaminated with organic chemicals is an urgent challenge. Most laboratories need to develop more efficient techniques to treat their wastewater; even if identified and labeled, it can decompose or react with other components of the matrix to produce a much more toxic compound. Phenol and its derivatives are very dangerous pollutants and once dissolved in water, they will be difficult to treat. Among the chemical indicators, phenolphthalein and methyl orange are well-known for their roles as endpoint indicators during acid-base titration due to their continuous and frequent use during research studies in the laboratory or practical [3]. There are several physical, chemical, and biological methods to treat and eliminate polluted effluents, including coagulation and flocculation, biodegradation, membrane filtration, chemical oxidation, ozonation, ion exchange, electrochemical treatment, and adsorption [4]. The most favorable method is adsorption; it has become an analytical method of choice, very efficient and simple in its use. Adsorption is commonly used in industry to treat water containing phenolic compounds. Activated carbon and natural clay are the most effective adsorbents, particularly recommended for treating dilute solutions [5].

Given that basic-colored indicators have a high capacity to dissolve in water, adsorption is considered to

be one of the most common and effective methods of treating the resulting pollution. The common use of phenolphthalein is as an indicator in acid-base titrations. It also serves as a universal indicator component, with methyl red, bromothymol blue, and thymol blue [6]. Phenolphthalein has been used as a laxative for over a century but is now removed from over-the-counter laxatives due to carcinogenicity concerns [7]. This research studies the adsorption of phenolphthalein on the surface of two different materials: activated carbon and natural clay. We report on how the adsorption process is affected by changes in the concentration of the colored indicator, the mass of the adsorbent, temperature, pH, and contact time, thereby examining kinetics as a function of adsorption isotherms for the Freundlich and Langmuir models.

■ EXPERIMENTAL SECTION

Materials

The materials used were natural clay (100%), activated carbon (98%), NaOH (100%), HCl (37%), ethanol (95%), phenolphthalein dye (99.92%) purchased from Merck (Germany), and double-distilled water.

Instrumentation

The instruments used in this study included an electronic balance, oven, UV-vis spectrophotometer (Thermo Genesys 10S), a magnetic stirrer, and a mercury thermometer. The adsorbents were characterized prior to adsorption by Fourier transform infrared spectroscopy (FTIR, Shimadzu 8000) and X-ray diffraction using an X'pert Pro diffractometer.

Procedure

Adsorption preparation

Both adsorbents were prepared from samples of activated carbon (coal 100) and natural clay (montmorillonite). The clay was first prepared from an aqueous dispersion, with continuous stirring for about 20 min at room temperature. The dispersion was then allowed to stand for 24 h and then dried in an oven at 110 °C for 24 h. Then the activated carbon was also dried at 110 °C for 24 h. The samples were pulverized and standardized using a sieve (38–300 nm), classified as ultrafine.

Process of adsorption

This study conducted the adsorption process using the batch method. The experiments involved varying contact times (2, 4, 6, 8, 24 h), phenolphthalein concentrations (50, 100, 150, 250, 500 mg L^{-1}), and pH levels of the phenolphthalein solution (4, 6, and 8). The sample was stirred using a stirrer at 300 rpm and then separated by filtration to isolate the two adsorbents. In addition, a UV-vis spectrophotometer was used to evaluate the phenolphthalein concentration of the solution after the adsorption process [8]. The process of adsorption is shown in Fig. 1. The analysis was carried out at 512 nm. The experiments were carried out with three replicates. Eq. (1–3) were used to calculate the percentage of phenolphthalein (%adsorption) and the quantity adsorbed (q_e);

$$q_e = (C_0 - C_t) \times \frac{V}{M} \quad (1)$$

$$\%T = \frac{C_i - C_e}{C_i} \times 100\% \quad (2)$$

$$C_e = C_0 - C_t \quad (3)$$

where q_e = amount of adsorbed dye (mg g^{-1}), C_i = initial phenolphthalein concentration before adsorption (mg L^{-1}), C_e = final phenolphthalein concentration after adsorption (mg L^{-1}), V = volume of phenolphthalein solution (L) and M = mass of clay and activated carbon (g).

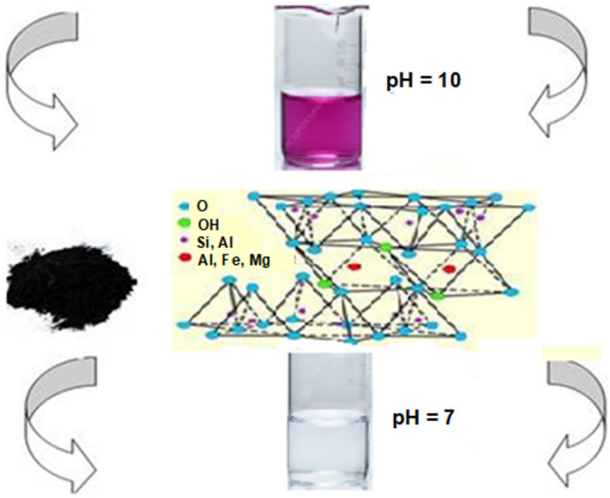

Fig 1. Adsorption process of phenolphthalein

■ RESULTS AND DISCUSSION

Fig. 2 shows the diffractogram of natural clay and activated carbon. According to the XRD diagram obtained, the clay has a mineralogical composition and inter-layer distances practically identical to natural clay [9]. The natural clay is characterized by 4 peaks. The first peak is located at 15.037 Å (001), and the other three are at 4.479 Å (110), 2.567 Å (200), and 1.498 Å (001).This diffractogram shows that the non-clay minerals present in varying amounts from one sample to another are mainly quartz with characteristic reflections at *d*001 = 3.35 and 4.28 Å, calcite (*d*001 = 3.21 Å), and feldspars (*d*00 = 4.06 Å) [10]. The XRD diffractogram of the activated carbon shows the presence of a large peak (2θ = 32°), indicating an amorphous structure.

FTIR Analysis

The FTIR tests confirmed that the analyzed materials have a crystalline structure. The spectra obtained are illustrated in Fig. 3. The band that spans between 1600 and 1700 cm^{-1} may be attributed to the valence vibrations of the OH group of the constituent water, in addition to the binding vibrations of the adsorbed water located at 1646 cm^{-1}. An absorption band centered on 3620 cm^{-1} is due to the valence vibrations of the OH groups bound to the octahedral Al cations (Al–O–Al). The Si–O bond is characterized by the intense band located between 900–1200 cm^{-1} and centered around 1009 cm^{-1}, which corresponds to the valence vibrations of the Si–O bond [11]. The bands between 795 and 748 cm^{-1}, coming from the Si–O–Al bond, also give way to a band around 778.4 cm^{-1}.

The FTIR spectrum of the compound phenolphthalein confirmed that this product has a crystalline structure. The band extends between 500 and 1500 cm^{-1}. Twenty absorption bands were found, all of the monomer molecules in solution, most of which can be attributed to well-established phenyl group frequencies [12]. In addition, carboxyl and quinonic groups (C–O) should absorb around 1680 cm^{-1}. The characteristic intense absorption associated with the carboxyl group is between 3500 and 2100 cm^{-1}. The results (Table 1) shows that the various components of the chemical analysis of natural clay. In this region, absorption does not occur at frequencies lower than 3100 cm^{-1}; there is little absorption beyond the C–H aromatic bands.

Table 1. Chemical composition of natural clay

Elements	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO
%	54.92	16.92	1.95	0.02	4.29
Elements	CaO	Na_2O	K_2O	TiO_2	P_2O_5
%	0.71	1.23	0.73	0.05	0.13

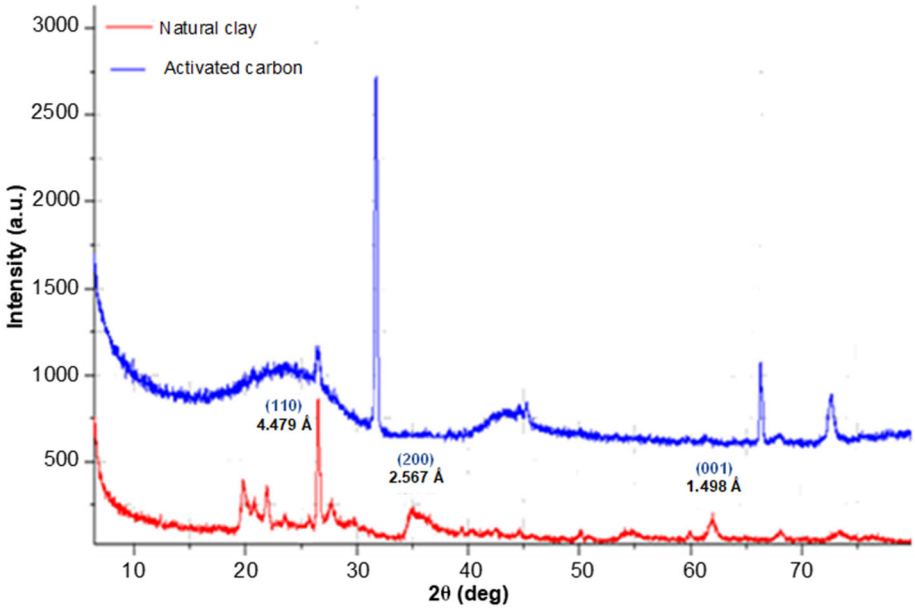

Fig 2. XRD diagram of natural clay and activated carbon

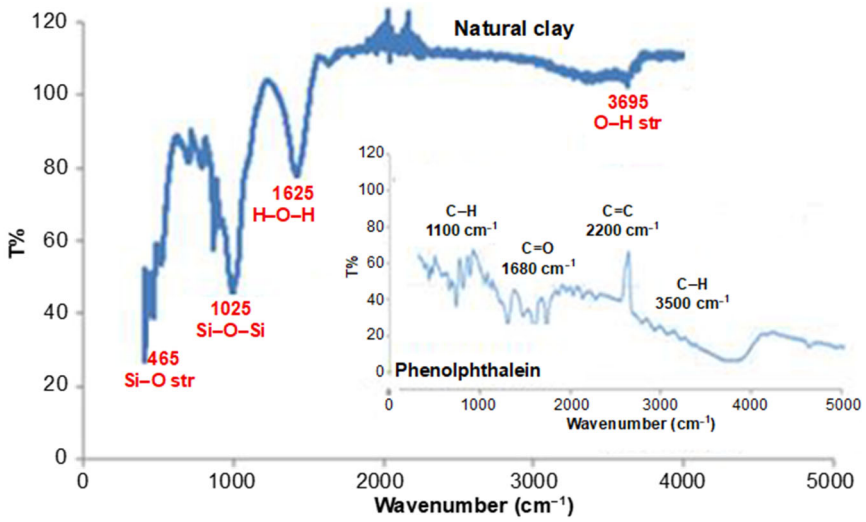

Fig 3. FTIR spectra for natural clay and phenolphthalein

Adsorption of Natural Clay and Activated Carbon on Phenolphthalein

It is common that agglomeration of a large number of molecular species at the liquid or solid phase surface occurred in relation to the mass. The development of adsorption is due to unbalanced or residual forces on the surface of the liquid or solid phase. The results in Fig. 4 show an increase in uptake concentrations in the gross clay curve, from 0.0005 to 0.0015 mg/L, followed by a decrease in uptake between the two concentrations of 0.0015and 0.0018 mg/g. This phenomenon results in physical adsorption of the type of adsorption isotherm II, which is the isotherm most commonly encountered when adsorption occurs on non-porous powders or those with macrospores (diameter greater than 500 Å). The point of inflection of the isotherms indicates that the first layer is fully saturated. Then, there is an increase in absorption up to that point.

It took the highest concentration of 0.0032 mg/g. For the carbon adsorption curve, there is a direct increase in absorption that corresponds to the increase in concentration, specific to an adsorbent with a particularly high microporous volume. Adsorption sites are usually saturated at low concentrations [13]. The form of the isotherm is characterized by a long plateau with a weak form of multilayer. This form is similar to the Langmuir model, which will be discussed later. In this case, the adsorption sites are considered to be equivalent.

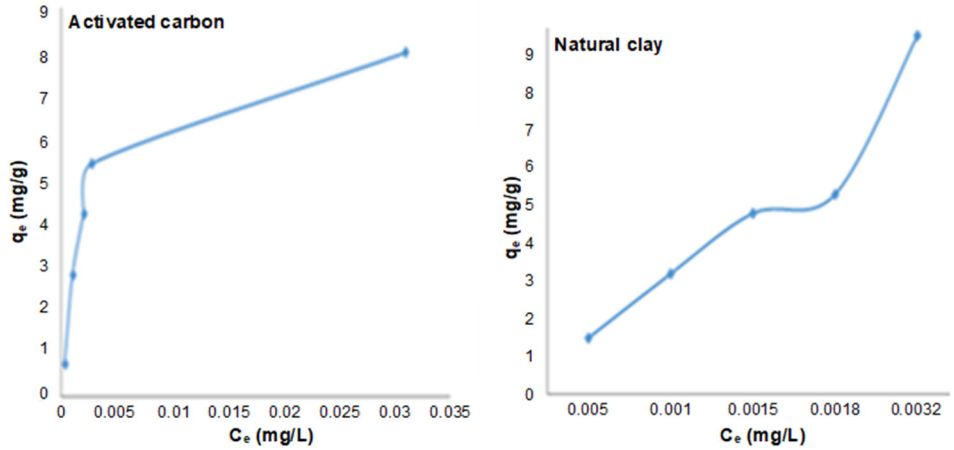

Fig 4. Initial concentration effect

Effect of Contact Time

To measure the effect of contact time, the solutions were prepared in the same way as before, using a fixed mass of activated carbon and natural clay (m = 100 mg) with varying concentrations of phenolphthalein at constant pH and temperature. Measurements were taken at different time periods (2, 4, 6, 8, 24 h), where the lowest concentration corresponds to the shortest time period, and the same applies to the clay. After placing the samples in a centrifuge, filtering and measuring the absorbance, the obtained results are shown in Fig. 5. With the increase in contact time, the adsorption capacity increases. There is also a similarity in adsorption capacity between activated charcoal and natural clay in the low concentration range and with the colored indicator, but the difference between them appears in the range of higher concentrations and longer contact time. This is because the adsorption process does not occur immediately. In porous materials, adsorption first occurs on available sites and on the external surface, then on the surface of internal pores, which takes longer.

Effect of Changing pH

A fixed mass of activated carbon (100 mg) was placed in 5 mL of the phenolphthalein solution at 30 mg/L, at a constant temperature and time, and at different pH values (4, 6, and 8). The same procedure is applied for clay. After that, the samples were placed in a centrifuge, filtered, and the absorbance was measured. The result obtained is shown in Fig. 6. In the case of natural clay, an increase in adsorption capacity can be observed with an increase in the

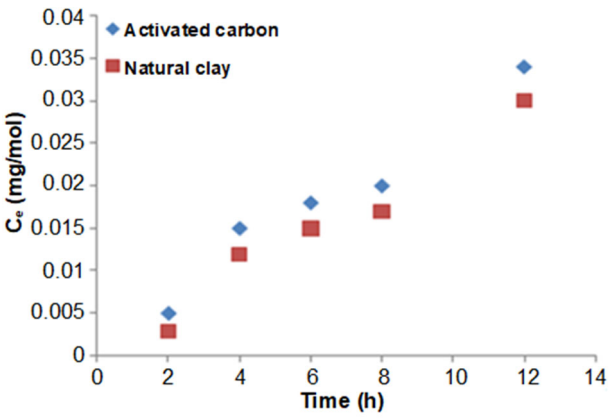

Fig 5. Effect of contact time

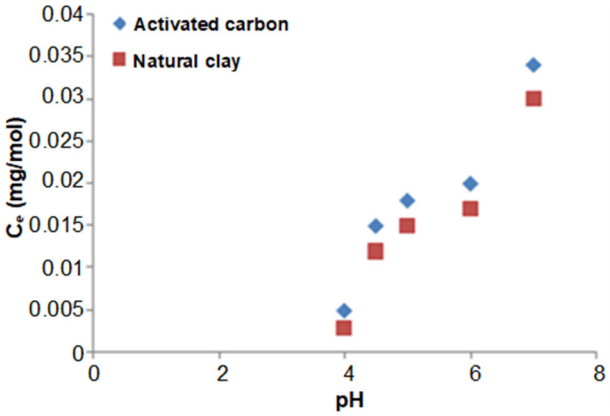

Fig 6. The effect of changing the pH

pH value, which is in the range 5 < pH < 8, and then tends to be fixed at a value close to 30 mg/g in the moderate base character range, As in the case of activated carbon, the adsorption capacity appears to be more stable with the change in pH because the value of the adsorption capacity in the acid, moderate and base ranges is close to 30 mg/g, with a slight increase in the acidic range which gives it an acidic character. Also, there is a gap in the adsorption capacity between activated carbon and natural clay. The low adsorption capacity in the acid field is due to the presence of a large number of H^+ ions, which compete with phenolphthalein molecules for adsorption sites [14]. In addition, clay is more affected by these ions because of its properties that differ from those of activated carbon in terms of the nature of the charges on its surface [15]. The absence of this effect is noted by the presence of H^+ ions in the moderate and basic range, where the adsorption capacity is large and more stable.

Effect of Changing Temperature

To measure the effect of temperature on the adsorption process, standard solutions with varying concentrations and different masses were prepared, totaling 5 mL for each solution. The lowest mass of carbon corresponded to the lowest concentration of the dye solution. The previous solutions were set at different temperatures (25, 30, 35, 40, 45 °C), where the lowest temperature corresponds to the lowest concentration, and the same applies to the natural clay. After that, the samples were placed in a centrifuge, filtered, and the

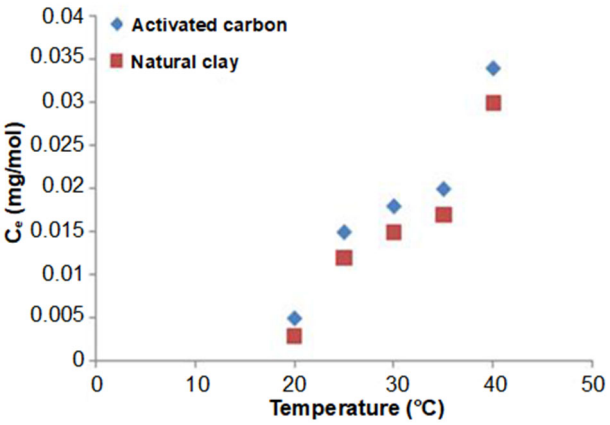

Fig 7. The effect of changing the temperature

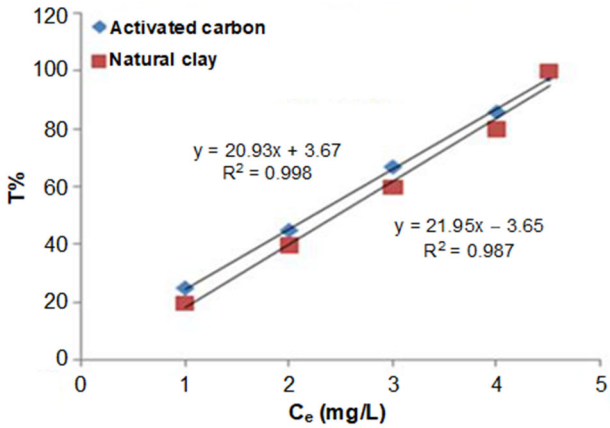

Fig 8. The effect of decolorization

absorbance was measured. Fig. 7 shows that the adsorption capacity decreases with increasing temperature, ranging from 22 mg/g at 25 °C to 11 mg/g at 45 °C for natural clay. As for activated carbon, the maximum adsorption capacity was 45 mg/g at 30 °C, reaching 35 mg/g at 45 °C. Notably, there is a discrepancy in the adsorption capacity of activated carbon and natural clay, as it is larger in carbon compared to clay. The reason for the decrease in adsorption capacity with increasing temperature is that the adsorption process is exothermic. Additionally, molecules at high temperatures possess high kinetic energy [16]. This reduces the chance of them collecting on the surface of the adsorbent material.

Decolorization Ratio (T%)

To identify the most efficient adsorbent material for removing color, the percentage of color removal for both activated carbon and natural clay was calculated and then compared. The same experimental data were used from the study that examined the effect of initial dye concentration, in which a fixed mass of adsorbent was tested with varying dye concentrations (Fig. 8).In the previous section, Fig. 5 represents the removal efficiency of phenolphthalein on a mass of two adsorbents in contact with a volume of 5 mL for a period of time (2, 4, 6, 8, 10 h). Several observations can be made: its effectiveness in removing the dye increases with increasing concentration to 97.5% at the highest concentration. As far as clay is concerned, it can be observed that its efficiency in removing the dye increases with concentration, reaching 99.7%. It can also be noted that the curve is divided into two parts: the range of minimum concentrations, which ends at a value close to 200 mg/L, and the other range is the range of major concentrations, starting from the previous value and ending at the maximum concentration of 500 mg/L. The bleaching efficiency of clay is high compared to that of activated carbon. This is due to the large interfacial surface area of the natural clay and its basic character, which allows it to absorb a large amount of dye. In contrast, activated charcoal has an acidic character so it absorbs a smaller amount of dye. The effectiveness of clay in removing color may be due to charges on its surface that cause ion exchange with dye molecules [17]. This quickly depletes its efficiency with high concentrations.

Adsorption Isotherms

Adsorption isotherms help to determine the nature of adsorption. For this purpose, solutions at varying concentrations (50 to 500 mg/L) with a variable adsorbent mass were used. After obtaining the results, a graph is drawn from Eq. (4) and (5).

$$\log q_e = f(\log C_e) \quad (4)$$

$$1/q_e = f(1/C_e) \quad (5)$$

The curves help to get the values of the constants of the two adsorption isotherms using the Freundlich and Langmuir equations (Eq. (6) and (7));

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \quad (6)$$

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{K_L} \times \frac{1}{q_m} \times \frac{1}{C_e} \quad (7)$$

where C_e: is equilibrium solute residual concentration (mg/L); q_m: is a maximum adsorption capacity (mg/g); K_L: is the adsorption equilibrium constant for the solute/adsorbent pair (mg/L); K_F and n are constants characteristic of the efficiency of an adsorbent with respect to a given solute.

The adsorption isotherm is a simple yet crucial tool in understanding adsorption mechanisms and assessing the best adsorbent for widespread applications. These isotherms provide information on the adsorbent/adsorbate affinity and an information of the energy of the links between the adsorbate and the adsorbent. Phenolphthalein adsorption has been studied according to the initial concentration of the dye. The results obtained were modeled using the two empirical models of Freundlich and Langmuir, as shown in Fig. 9 and 10.

The Freundlich model predicts that the concentration of coloring on the adsorbent will increase as long as the concentration of coloring in the liquid phase increases. However, experimental evidence indicates that an isothermal platform is achieved at a limit value of the solid phase concentration. This tray is not predicted by the Freundlich equation. Consequently, the equation has no meaning in the real physical phenomenon [18].

Langmuir's adsorption isotherm is used to describe the balance between the adsorbent and the adsorption system, where adsorption is limited to a molecular layer

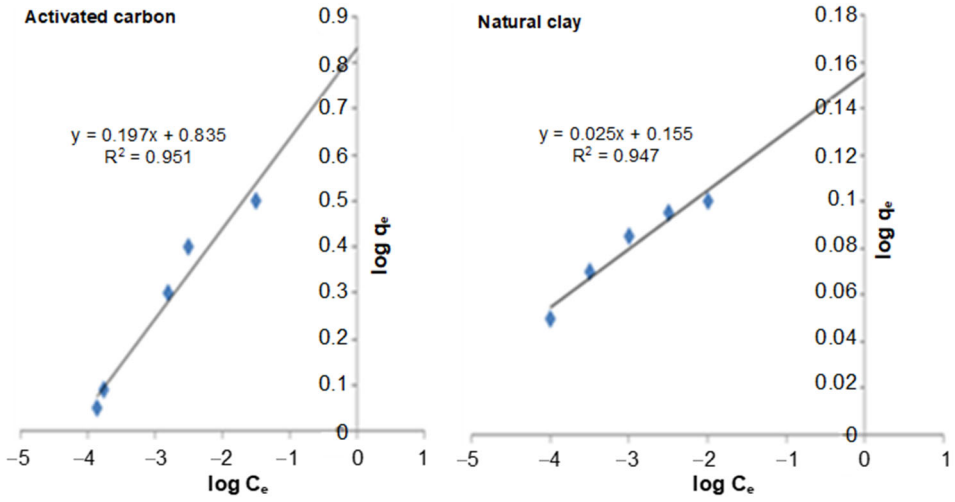

Fig 9. Freundlich isotherm model

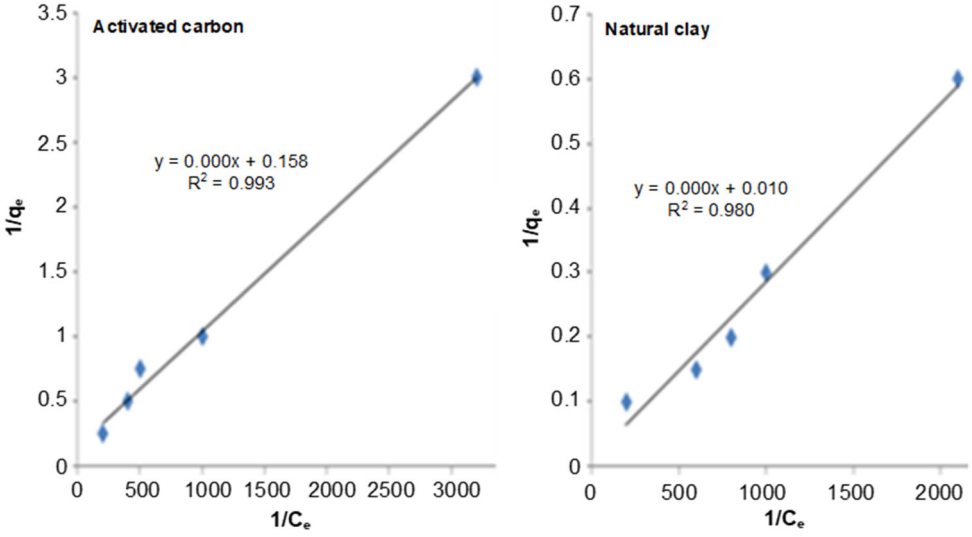

Fig 10. Langmuir isotherm model

with or before relative unit pressure. Although the isotherm initially proposed by Langmuir in 1918, it is generally appropriate to describe the chemisorption process when ionic or covalent chemical bonds are formed between adsorbents and adsorbents [19]. Langmuir's adsorption isotherm is based on the hypothesis of a structurally homogeneous adsorbent, where all sorption sites are identical and energetically equivalent. Therefore, the sorbent has a finite capacity for the sorbate. These curves can be used to obtain the constants in Table 2. The Langmuir isotherm equation may be expressed in a linearized form as shown in Eq. (7).

After calculating the correlation coefficients and constants of the Freundlich and Langmuir equations, in both cases, for the Freundlich or Langmuir isotherm and for activated carbon or natural clay, the two previous models do not fully describe the adsorption (based on correlation coefficients), which may be justified for the Freundlich isotherm [21], because it describes very precisely adsorption at low concentrations, while it fails in large concentrations. In any case, it is possible to determine which of the two models is most suitable for describing adsorption. For instance, in the case of clay, the correlation coefficient for the Freundlich model is 0.947, whereas for the Langmuir model, it is 0.980. This indicates that the Langmuir model is more suitable for describing the adsorption of phenolphthalein on clay, suggesting that the adsorption is multilayer or physical, which characterizes irregular surfaces.

The Freundlich isotherm is only applicable to the low adsorption pressure region. It is important to keep these limitations in mind when interpreting results obtained by fitting the Freundlich isotherm to experimental data [22]. Meanwhile, Langmuir adsorption isotherms assume that the adsorption is a monolayer. However, this formation is also only possible at low pressure [23].

In this study, the Freundlich and the Langmuir isotherm models were used to analyze the equilibrium data. The adsorption capacity, type of coverage, and rate constants can be determined using these adsorption isotherm models. The R^2 values were used to determine which model was more applicable. Fig. 11 shows the data for activated carbon, where the correlation coefficient for the Freundlich model is 0.980, whereas for the Langmuir model, it is 0.993 [24]. This means that the Langmuir model is more suitable for describing the adsorption of phenolphthalein on activated carbon, as this model describes single-ply adsorption, which characterizes homogeneous surfaces.

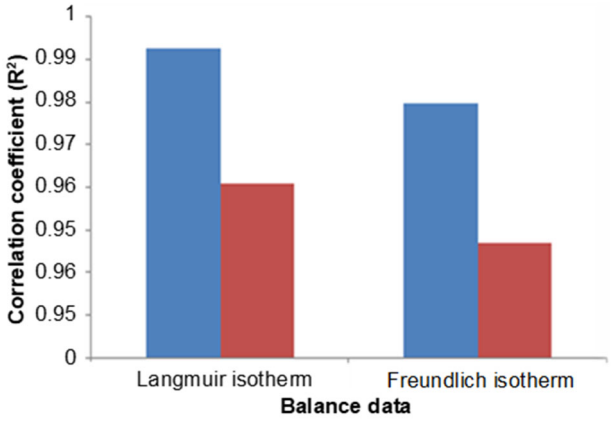

Fig 11. Effect of the number on the correlation coefficient (R^2)

Table 2. Isotherm parameters obtained from the fit of various equations

Isotherm models	Activated carbon	Natural clay	Heydari et al. [6]	Tetteh et al. [20]
Langmuir				
q_e (mg/g)	31 ± 0.20	30 ± 0.01	41± 0.01	24.80 ± 0.10
K_L (L/mg)	0.1 ± 0.10	0.040 ± 0.05	0.069 ± 0.10	1.04 ± 0.10
R^2	0.993	0.980	0.948	0.859
Freundlich				
K_F (mg/g)(L $mg^{-1})^{1/n}$	30.00 ± 0.10	29.00 ± 0.10	35.77	35.77
n	0.99 ± 0.10	4.04 ± 0.10	1.40 ± 0.01	2.23 ± 0.01
R^2	0.961	0.947	0.987	0.850

■ CONCLUSION

Based on the results, this study demonstrates that the phenolphthalein adsorption process using natural clay is more efficient than that using activated carbon in the high dye concentration range. The maximum adsorption capacity obtained was 31 ± 0.2 mg/g under the following conditions: a contact time of 120 min, 100 mg of natural clay and activated carbon, a solution pH of 6, and an initial phenolphthalein concentration of 30 mg/L. The physicochemical parameters are essential for facilitating interactions with phenolphthalein and the dye adsorbent functional groups, with a q_e of 31 mg/g, a K_L of 0.1. The Freundlich model is more suitable for clay, while the Langmuir model is more suitable for activated carbon. The results confirm that natural clay and activated carbon are effective alternatives for removing phenolphthalein from water. The adsorption process is spontaneous, endothermic, and characterized by medium confusion. These results suggest that the adsorption of dyes by raw clay and activated carbon may be due to physisorption. Finally, the use of natural Algerian clay has greater potential for removing laboratory dyes, as it does not require expensive equipment.

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■ CONFLICT OF INTEREST

The authors have no conflict of interest.

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