An organified mixture of illite-kaolinite for the removal of Congo red from wastewater

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ABSTRACT
Congo red (CR), an anionic dye, is very harmful to the environment and should be removed via an easy and inexpensive technique. The adsorption of CR from an aqueous solution onto a mixture of illite-kaolinite clay minerals and surfactant-modified illite-kaolinite was investigated at different temperatures. The characterization of the raw clay mixture and the organified sample was confirmed with X-ray diffraction, FTIR spectroscopy, nitrogen gas adsorption at 77 K and transmission electronic microscopy techniques. Factors influencing the adsorption such as pH, adsorbent dosage and temperature were tested. The thermodynamic parameters $\Delta G$, $\Delta H$ and $\Delta S$ have been calculated, and it was found that the adsorption is exothermic in nature. The isotherm data were fitted by the linear form of the Langmuir and Freundlich models and showed a good fit with the Langmuir approach. The maximum adsorption capacity of surfactant-modified clay (83 mg/g) was found to be around 14 times higher than that of unmodified clay. Kinetic studies of CR on illite-kaolinite and organified illite-kaolinite were evaluated by pseudo-first-order and pseudo-second-order models.

1. Introduction

Congo Red (CR) was introduced in Germany in 1885 as an extremely valuable textile dye, especially for cotton. Additionally, CR is employed in paper, leather and plastic industries [1,2]. This compound was the first of many so-called direct dyes. CR has a complex chemical structure with high solubility in aqueous solutions and a highly persistent nature once it has been discharged in the environment. Due to its carcinogenic effects on humans, CR has been banned in many countries [3]. However, several developing countries that produce cotton still use CR in their textile industries. For example, in Sudan, approximately 37.5 thousand tons of CR-dyed cotton are produced every year. This growing demand for cotton gives rise to an important amount of wastewater contaminated with this toxic dye; hence, there is a need for this issue to be treated.

Clay minerals exhibit a great potential and prospective as a cost-efficient adsorbent. Many studies have been carried out on the removal of cationic dyes using natural clay minerals [4–6]. Their results showed that cationic dyes such as methylene blue (MB) and crystal violet (CV) are more adsorbed by natural clay minerals than are the anionic ones such as CR dye. The adsorbed amounts, $q_m$, on illite-kaolinite are 98.06, 82.13 and 23.52 mg/g for MB, CV and CR respectively. The low adsorption capacity towards anionic dyes is attributed to the electrostatic repulsion between CR and clay’s negatively charged surface [7]. Hence, the adsorbent surface should be modified to enhance its adsorption capacity. Several methods can be used to modify the surface of clay mineral, such as calcination processes [8], acid treatment [9] or, recently organoclay formation [10,11]. The surface properties of leonardite, a shale clay rich in humic and fulvic acids, change by carbonization at 900°C, causing an increase in the adsorption capacity of this clay to CR [12]. Acid treatment of kaolinite and montmorillonite enhances their capacity of adsorption of CR. After acid treatment, the maximum adsorption of this dye increases by 50% for kaolinite and by 15% for montmorillonite [13]. For example, the maximum amount of CR adsorbed at room temperature onto three Australian kaolins, 1:1 clay, varies between 5.58 and 6.89 mg/g [14]. Additionally, the maximum adsorbed amount of CR on the purified montmorillonite is 10.2 mg/g.

On the other hand, organo-attapulgite shows a high adsorption capacity for CR (189.39 mg/g) from an aqueous solution at room temperature [7]. Surfactant-modified montmorillonite displays a higher adsorption...
capacity for CR than untreated montmorillonite does, $q_e = 127 \text{ mg/g}$ [15]. The adsorption capacities of two organophilic hectorites for CR are in the range of 180 and 200 mg/g at different temperatures between 303 and 333 K, whereas the dye removal percentage never exceeds 10% for the parent hectorite, which is 2:1 smectite clay [16]. The effectiveness of organo-vermiculite to uptake CR dye from an aqueous solution at 298 K is 73 times higher than that of the parent vermiculite [17]. Similar results were also obtained for the removal of CR from the aqueous solution using organo-rectorite [18]. In Table 1, we report the following data from the literature for the adsorption of CR onto organophilic clay. From this literature review, it is evident that organoclays show higher efficiencies for CR removal from aqueous solutions than raw clays do.

Currently, the composition of wastewater is becoming more complex. The composition includes different types of contaminants (heavy metals and organic dyes). Among these organic dyes, MB, CV and CR are the most harmful to health and the environment [5,19]. A previous study [4] showed the possibility for the illite-kaolinite clay minerals to remove MB and CV from wastewater. To the best of our knowledge, few studies have been focused on using natural mixture clay minerals to remove anionic dyes from wastewater. Our present investigation is one in a series of studies devoted to the removal of cationic and anionic dyes from wastewater using a natural mixture of clay minerals. The mixture of illite-kaolinite clay minerals was modified with didecyldimethylammonium bromide (cationic surfactant) to study the kinetics and thermodynamics of CR adsorption.

2. Materials and method

2.1. Clay minerals

Illite-kaolinite clay minerals (IKAol) used in this study were supplied from Almarwani For Spices Company, KSA. The raw natural mixture is saturated with sodium chloride to eliminate cations from the interlayer space according to the ion exchange process before use [24]. The sodic clay is gently ground, and the fine fraction of particles less than 100 μm is used in the following experiments.

2.2. Chemicals

The surfactant didecyldimethylammonium bromide ([(CH$_3$)(CH$_2$)$_9$]$_2$N(CH$_3$)$_2$Br) and CR (C.I. 30025, Chemical formula, C$_{32}$H$_{22}$N$_6$Na$_2$O$_6$S$_2$, $M = 696.69$) are from Sigma-Aldrich. The chemical structure of the surfactant and Congo red are given in Figure 1.

The surfactant and the dye are used without any purification, and all the other chemicals used are analytical-reagent grade. All solutions are prepared with bi-distilled water.

2.3. Preparation of organophilic illite-kaolinite

The preparation of the organified illite-kaolinite is performed as follows: 10 g of Na-clay is dispersed in 250 ml of bi-distilled water and stirred for an hour at 70°C in a water bath, to which an adequate amount of a prepared solution of didecyldimethylammonium bromide (DDAB) is slowly added [25,26]. The mixture is kept under magnetic agitation for 8 h at 70°C. Then, the

| Organophilic clay | Adsorption capacity ($q_{max}$) (mg/g) | Isotherm study | Kinetic study | Thermodynamic study | Equilibrium conditions: pH | Dosage of clay | References |
|-------------------|---------------------------------------|----------------|--------------|---------------------|---------------------------|----------------|------------|
| Cetyltrimethylammonium bromide-modified Bentonite | 210 | Langmuir | Pseudo-second order | Endothermic and spontaneous in nature | 5–9, 120 min | 2 g/L | [20] |
| Cetyltrimethylammonium bromide-modified Kaolin | 24.46 | Langmuir | Pseudo-second order | —— | —— | 2 g/L | [21] |
| Cetyltrimethylammonium bromide-modified Montmorillonite | 351.00 | Langmuir | Pseudo-second order | Endothermic | 7.5, 1–100 min | 2 g/L | [18] |
| Cetyltrimethylammonium bromide-modified Rectorite | 153.85 | Langmuir | Pseudo-second order | Endothermic and spontaneous in nature | 7, 240 min | 1.0 g/L | [18] |
| Hexadecyltrimethylammonium bromide-modified Attapulgite | 189.39 | Langmuir | Pseudo-second order | Endothermic | 4, 120 min | 2 g/L | [7] |
| Hexadecyltrimethylammonium bromide-modified Bentonite | 16.04 | Langmuir | Pseudo-second order | Endothermic and spontaneous in nature | 5–9, 15–90 min | 0.1 g/L | [22] |
| Hexadecyltrimethylammonium bromide-modified Vermiculite | 192.31 | Langmuir | Pseudo-second order | Endothermic and spontaneous in nature | ——, 24 h | 1.0 g/L | [22] |
| Dodecyltrimethylammonium bromide-modified Montmorillonite | 83.60 | —— | —— | —— | —— | 2 g/L | [15] |
| 1,3-bis(dodecyldimethylammonio)-2-hydroxypropane dichloride-modified Montmorillonite | 192.57 | Langmuir | Pseudo-second order | Endothermic and spontaneous in nature | 7, 90–120 min | 1.6 g/L | [23] |
suspended solid is filtered and washed several times with bi-distilled water until the silver nitrate test for halogen ions is shown to be negative. The organophilic clay (DDAB-IKaol) is freeze-dried and ground with a mortar to obtain a fine texture.

### 2.4. Characterization of samples

The X-ray diffractograms of the parent and organified clay are obtained on a Phillips Xpert-pro diffractometer using Cu Kα radiation (λ = 1.54060 Å), and 2θ varies from 5° to 60°. FTIR spectra in the 4000–400 cm⁻¹ region are measured on a Thermo Scientific spectrometer with a deuterated triglycine sulfate (DTGS) detector. The KBr pellet method is used. A Micromeritics ASAP2000 automatic apparatus (Micromeritics, Norcross, GA) is used to determine the specific surface areas and pore size distribution according to the adsorption–desorption isotherms of N₂ at 77 K. Finally, transmission electron microscope image analysis of the adsorbent without and with a surfactant are examined on a TEM (JEOL JEM 1400 series) at 120 kV.

#### 2.5. Batch adsorption experiments

The experimental procedure is identical to that described in our earlier work [4,6]. The amount of CR adsorbed onto IKaol and DDAB-IKaol is determined by the difference between the initial and remaining concentration of CR solution by the following expression:

\[
q_e = \frac{(C_0 - C_e)V}{m}
\]

where \(q_e\) is the amount of dye adsorbed per unit weight of the organoclay in (mg/g), \(V\) is the volume of solution in (L), \(C_0\) is the initial metal ion concentration in (mg/L), \(C_e\) is the equilibrium CR concentration in (mg/L) and \(m\) is the dry weight of the organophilic clay in (g).

### 3. Results and discussion

#### 3.1. Characterization of adsorbents IKaol and DDAB-IKaol

##### 3.1.1. XRD analysis

The XRD patterns of the IKaol sample and DDAB-IKaol are depicted in Figure 2. This figure shows that there is no change on the basal spacing due to the insertion of the surfactant into the interlayer space of the clay.

##### 3.1.2. FTIR analysis

The FTIR spectra of IK and organoclay (DDAB-IKaol) are shown in Figure 3. It can be seen from this figure that those bands related to raw clay remained nearly unchanged after the organic modifications. Only the bands attributed to the stretching vibration of \(-\text{CH}_2\) (2850 cm⁻¹) and \(-\text{CH}_3\) (2925 cm⁻¹) are observed on the spectra of organoclay. The band observed at 1395 cm⁻¹ is attributed to the C–N group of the surfactant [21]. This band indicates the intercalation of surfactant molecules between silicates. No shifting is observed for the position of the weak band observed at 1631 cm⁻¹, which is attributed to the deformation mode of the interlayer water molecules (δ H-O-H).

##### 3.1.3. Surface area measurement

Figure 4 exhibits the N₂ adsorption–desorption isotherms of illite-kaolinite (IKaol) clay minerals and DDAB-IKaol. This figure shows that the adsorption–desorption isotherms did not preserve overlap, increasing the lag phenomenon of regress and presenting a type H3 hysteresis loop, which is characteristic of interstitial pores.

Table 2 reports the textural properties of the two adsorbents used (IKaol and DDAB-IKaol). The decrease in the surface area is due to the large number of active sites satisfied by surfactant molecules and the reduction of the accessible internal surface for nitrogen gas. However, the organoclay (DDAB-IKaol) exhibits a larger pore size than that of the raw clay. The same result was observed for the organified rectorite, and this order of magnitude difference in the pore size may match well with the size of the CR molecule, favouring its removal [18].

##### 3.1.4. TEM image analysis

Figure 5 shows the TEM images of illite-kaolinite (IKaol) and DDAB-IKaol clay. From this figure, typical and well-ordered arrangements of clay mineral platelets are shown, and stacks of multilayers become more dispersive after modification with the surfactant. It can also be seen that the distance from layer to layer is much larger and more apparent in the case of the organified clay. A similar behavior has been reported in the literature [27].
Figure 2. X-ray diffraction (XRD) patterns of illite-kaolinite (a) unmodified (b) with surfactant.

Figure 3. FTIR spectra (Dotted line for raw clay IKaol and continuous line for organified clay DDAB-IKaol).

Figure 4. $N_2$ adsorption-desorption isotherms of samples (A) IKaol, (B) DDAB-IKaol.
Table 2. The textural properties of the raw and organoclay determined from nitrogen adsorption/desorption isotherms.

| Adsorbent | \( S_{\text{BET}} \) (m\(^2\)/g) | \( S_{\text{BJH}} \) (m\(^2\)/g) | Total pore volume (cm\(^3\)/g) | Average pore size by BET (Å) |
|-----------|---------------------------------|---------------------------------|-------------------------------|-----------------------------|
| Raw clay  | 128                             | 86                              | 0.320                         | 75.6                        |
| Organoclay | 39                              | 40                              | 0.181                         | 125.8                       |

Figure 5. TEM images of IKaol and DDAB-IKaol samples.

3.2. Isotherm and kinetic model results

3.2.1. Effect of pH
The data recorded in Figure 6 show that the influence of the pH on CR removal from aqueous solution using IKaol or DDAB-IKaol clay is not significant. In these two cases, when the pH increases from pH 6–11, the \( q_e \) decreases by approximately 10 mg/g. The minor decrease may be attributed to the high repulsion between acidic CR molecules and the hydroxide ion OH\(^-\) at the surface of clay, especially at a higher pH values.

3.2.2. Effect of adsorbent dosage
The effect of adsorbent dosage on the removal of CR by IKaol and DDAB-IKaol is shown in Figure 7. This figure indicates that the adsorption by DDAB-IKaol was enhanced rapidly by increasing the amount of organified clay from 0.2 to 1.8 g/L and was slightly enhanced for IKaol without surfactant.

3.3. Thermodynamic studies
Thermodynamic parameters of \( \Delta G^\circ \), \( \Delta H^\circ \) and \( \Delta S^\circ \) for the adsorption processes are calculated using the following equations:

\[
\Delta G^\circ = -RT \ln K_c
\]

\[
\Delta G^\circ = \Delta H^\circ - T \Delta S^\circ
\]

where \( R \) is the universal gas constant (8.314 J/mol.K), \( K_c \) is the Langmuir constant, and \( T \) is the absolute temperature in K. \( \Delta H^\circ \) and \( \Delta S^\circ \) could be calculated from the Van’t Hoff equation:

\[
\ln K_c = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT}
\]

The enthalpy changes and the entropy changes are obtained from the slope and the intercept of the plot \( \ln K_c \) vs \( 1/T \) (Figure 8), and these values are summarized in Table 3.
Figure 6. Effect of pH on adsorbed amount of CR onto IKaol and DDAB-IKaol (T = 293.15 K, equilibrium time = 180 min, [CR]₀ = 150 ppm, adsorbent dosage 0.6 g/L).

Figure 7. The effect of adsorbent dosage IKaol and DDAB-IKaol on CR removal (T = 293.15 K, equilibrium time = 180 min, [CR]₀ = 150 ppm).

Figure 8. The plot Ln Kc versus 1/T for CR adsorption on IKaol and DDAB-IKaol clay (equilibrium time = 180 min, pH = 6 and adsorbent dosage 0.6 g/L).
### Table 3. Thermodynamic parameters for adsorption of CR by IKaol and DDAB-IKaol at different temperatures.

| Sample       | \(\Delta S^0\) (J/mol.K) | \(\Delta H^0\) (kJ/mol) | 293.15 | 303.15 | 313.15 | 323.15 |
|--------------|--------------------------|--------------------------|--------|--------|--------|--------|
| IKaol        | -22.5 (-23)*             | -2.1                     | 4.56   | 4.78 (4.88)** | 5.01 (5.33)** | 5.23 (6.17)** |
| DDAB-IKaol   | -34.7                    | -7.9                     | 2.30   | 2.65   | 2.99   | 3.34   |

*from reference [6], b from reference [28]

### Table 4. Isotherm parameters obtained for CR adsorption onto IKaol and DDAB-IKaol clay at 293.15 K

| Adsorbent    | qmax (mg/g) | -kL (L/mg) | R²     | KF (L/mg) | n     | R²     |
|--------------|-------------|------------|--------|-----------|-------|--------|
| IKaol        | 5.7 (5.74)* | 0.0105     | 0.99   | 752.2     | -1.308| 0.83   |
| DDAB-IKaol   | 83.0        | 0.0100     | 0.99   | 12.28     | 3.540 | 0.97   |

*from reference [5].

The negative value of \(\Delta H^0\) indicates that the adsorption of CR is an exothermic process, while the positive values of \(\Delta G^0\) at various temperatures indicate that the process is nonspontaneous. These values are in the order of magnitude of those obtained for the adsorption of CR on natural serpentine [28].

### 3.4. Adsorption isotherms

To investigate the interaction between CR and IKaol or DDAB-IKaol, two kinds of isotherm equations (Langmuir model and Freundlich model) are tested to fit our experimental database. The Langmuir isotherm assumes that the adsorption of CR occurs in a monolayer, while the Freundlich isotherm assumes that the adsorption occurs on some heterogeneous surface and proceeds in a reversible way. The amount of the adsorbed CR (mg/g) at the solid/liquid interface, according to these two models, is given by the following equations:

- **Langmuir equation**
  \[
  q_e = \frac{k_L q_{max} C_e}{1 + k_L C_e}
  \]  

- **Freundlich equation**
  \[
  q_e = k_F C_e^{1/n}
  \]  

where \(q_{max}\) is the maximum adsorption capacity of the clay (mg/g), \(k_L\) (L/mg) is the Langmuir adsorption constant, \(k_F\) is the Freundlich constant and \(1/n\) is a factor relative to the adsorption intensity. The parameters evaluated by the two equations are summarized in Table 4.

The results obtained are shown in Figure 9. Based on the correlation coefficient values, it was possible to conclude that the Langmuir model \(R^2 = 0.97\) is a better fit for the experimental equilibrium data than the Freundlich isotherm \(R^2 = 0.83\), which indicates that the adsorption process was monolayer formation. The \(q_{max}\) calculated from the Langmuir isotherm is equal to 83 mg/g at 293.15 K and is coherent with the experimental value. The maximum adsorption of CR onto IKaol without a surfactant is comparable to the value of \(q_{max}\), corresponding to the adsorption of CR onto pure Kaolin mineral clay [5].

![Figure 9. Equilibrium isotherm plots for CR adsorption onto IKaol and DDAB-IKaol at 293.15 K (Langmuir model: solid lines; Freundlich model dashes lines).](image-url)
Table 5. Characteristic parameters of kinetic models

|                 | Pseudo-first-order model |       | Pseudo-second-order model |       |
|-----------------|--------------------------|-------|---------------------------|-------|
|                 | qₑ (mg/g)                | k₁    | R²                        | qₑ (mg/g) | k₂    | R² |
| IKaol           | 131.82                   | 9.21  | 0.73                      | 1.05     | 4.86  | 0.99|
| DDAB-IKaol      | 96.6                     | 16.12 | 0.95                      | 4.35     | 1.66  | 0.99|

Figure 10. Representation of the kinetic data of CR adsorption onto IKaol and DDAB-IKaol at 293.15 K, equilibrium time = 3 h, pH = 6 and adsorbent dosage 0.6 g/L.

Adsorption kinetics

To understand and describe the adsorption mechanisms, the pseudo-first-order and pseudo-second-order models, many models were applied. The following equation represents the linear form of the pseudo-first-order model:

\[
\log(qₑ - qₜ) = \log qₑ - \frac{k_1}{2.303} t
\]  (7)

A linear form of the pseudo-second-order kinetic model is given as:

\[
\frac{t}{qₜ} = \frac{1}{k_2 qₑ^2} + \frac{1}{qₑ} t
\]  (8)

The obtained results are listed in Table 5. The representation of the kinetic data according to the two models is given in Figure 10. A higher R² value (greater than 0.99) denoted better model fitting. Therefore, the pseudo-second-order model is the suitable approach for predicting the adsorption kinetics of CR on IKaol and on DDAB-IKaol.

4. Conclusion

In this work, a surfactant-modified mixture of natural clay (illite-kaolinite) was used as an adsorbent to remove CR from aqueous solution. The obtained results show that the organified clay has a maximum adsorption capacity equal to 83 mg/g, much higher than that of the unmodified mixture and consistent with experimental data. This may be attributed to the changes in the textural properties of DDAB-IKaol.

The results also indicated that all the adsorption processes were pseudo-second order and followed the Langmuir theory which implies that the adsorption of CR on IKaol and DDAB-IKaol was monolayer.

Moreover, the thermodynamic process was not affected by the modification of IKaol using cationic surfactant, and calculated parameters indicate that the CR adsorption onto the mixture (modified or unmodified) is exothermic and favourable at a lower temperature.

As a result, it can be said that the surfactant that modified the mixture of illite-kaolinite is an effective, less expensive adsorbent for the removal of CR from wastewater.

Disclosure statement

No potential conflict of interest was reported by the authors.

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References

[1] Omidi Khaniabadi Y, Basiri H, Nourmoradi H, et al. Adsorption of Congo Red Dye from aqueous solutions by montmorillonite as a Low-cost adsorbent. Int. J. Chem. React. Eng. 2018;16–21.
[2] Steensma DP. “Congo” red: out of Africa? Arch. Pathol. Lab. Med. 2001;125:250–252.
[3] Raval NP, Shah PU, Shah NK. Adsorptive amputation of hazardous azo dye Congo red from wastewater: a critical review. Environ. Sci. Pollut. Res. 2016;23:14810–14853.
[4] Sakin Omer O, Hussein MA, Hussein BHM, et al. Adsorption thermodynamics of cationic dyes (methylene blue and crystal violet) to a natural clay mineral from aqueous
solution between 293.15 and 323.15 K. Arab. J. Chem. 2018;11:615–623.

[5] Bentahar S, Dbik A, Khomri ME, et al. Adsorption of methylene blue, crystal violet and Congo red from binary and ternary systems with natural clay: kinetic, isotherm, and thermodynamic. J. Environ. Chem. Eng. 2017;5:5921–5932.

[6] Sakin Omer O, Hussein BHM, Hussein MA, et al. Mixture of illite-kaolinite for efficient water purification: removal of As(III) from aqueous solutions. Desalin. Water Treat. 2017;79:273–281.

[7] Chen H, Zhao J. Adsorption study for removal of Congo red anionic dye using organo-attapulgite. Adsorption. 2009;15:381–389.

[8] Chen H, Zhao J. Adsorption study for removal of Congo red anionic dye using organo-attapulgite. Adsorption. 2009;15:381–389.

[9] Komadel P. Acid activated clays: materials in continuous demand. Appl. Clay Sci. 2016;131:84–99.

[10] Zhu L, Wang L, Xu Y. Chitosan and surfactant co-modified montmorillonite: A multifunctional adsorbent for contaminant removal. Appl. Clay Sci. 2017;146:35–42.

[11] Kausar A, Iqbal M, Javed A, et al. Dyes adsorption using clay and modified clay: A review. J. Mol. Liq. 2018;256:395–407.

[12] Ausavasuhi A, Kampoosaen C, Kengnok O. Adsorption characteristics of Congo red on carbonized Leonardite. J. Clean. Prod. 2016;134:506–514.

[13] Bhattacharyya KG, Sen GS, Sarma GK. Kinetics, equilibrium isotherms and thermodynamics of adsorption of Congo red onto natural and acid-treated kaolinite and montmorillonite. Desalin. Water Treat. 2015;53:530–542.

[14] Vimonses V, Lei S, Jin B, et al. Adsorption of Congo red by three Australian kaolins. Appl. Clay Sci. 2009;43:465–472.

[15] Wang L, Wang A. Adsorption properties of Congo Red from aqueous solution on surfactant-modified montmorillonite. J. Hazard. Mater. 2008;160:173–180.

[16] Xia C, Jing Y, Jia Y, et al. Adsorption properties of Congo red from aqueous solution on modified hectorite: kinetic and thermodynamic studies. Desalination. 2011;265:81–87.

[17] Yu X, Wei C, Ke L, et al. Development of organovermiculite-based adsorbent for removing anionic dye from aqueous solution. J. Hazard. Mater. 2010;180:499–507.

[18] Liu Y, Wang W, Wang A. Removal of Congo red from aqueous solution by sorption on organified rectorite. Clean - soil, Air. Water (Basel). 2010;38:670–677.

[19] Bentahar S, Dbik A, El Khomri M, et al. Study of removal of Congo red by local natural clay. St. Circ. St. CICBIA. 2016;17:295–307.

[20] Youssef AM AM, Al-Awadhi MM. Adsorption of acid dyes onto bentonite and surfactant-modified bentonite. J. Anal. Bioanal. Tech. 2013;04:1–7.

[21] Zenasni MA, Meroufel B, Merlin A, et al. Adsorption of Congo Red from aqueous solution using CTAB-kaolin from bechar Algeria. J. Surf. Eng. Mater. Adv. Technol. 2014;04:332–341.

[22] Fosso-Kankeu E, Waanders F, Fournier CL. Adsorption of Congo Red by surfactant-impregnated bentonite clay. Desalin. Water Treat. 2016;57:1–9.

[23] Gu Z, Gao M, Luo Z, et al. Gemini surfactant modified montmorillonite as highly efficient adsorbent for anionic dyes. Sep. Sci. Technol. 2014;49:2878–2889.

[24] Bergaya F, Lagaly G. Purification of natural clays. Dev. Clay Sci. 1st ed. Elsevier Inc. 1st ed. Elsevier Inc. 2013:1:213–221.

[25] Park Y, Ayoko GA, Kurdi R, et al. Adsorption of phenolic compounds by organoclays: implications for the removal of organic pollutants from aqueous media. J. Colloid Interface Sci. 2013;406:196–208.

[26] He H, Ma Y, Zhu J, et al. Organoclays prepared from montmorillonites with different cation exchange capacity and surfactant configuration. Appl. Clay Sci. 2010;48:67–72.

[27] Wang L, Wang A. Adsorption properties of Congo Red from aqueous solution onto surfactant-modified montmorillonite. J. Hazard. Mater. 2008;160:173–180.

[28] Shaban M, Abu Khadra MR, Khan AAP, et al. Removal of Congo red, methylene blue and Cr(VI) ions from water using natural serpentine. J. Taiwan Inst. Chem. Eng. 2017;0:1–15.