Adsorption capability for Congo Red on exfoliated graphene-decorated CoFe$_2$O$_4$ nanocomposite: Kinetic, isotherm, thermodynamic and recyclability studies

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Abstract. In the present work, we investigated kinetics and isotherms of the adsorption process of EG@CoFe$_2$O$_4$ adsorbent toward Congo Red dye in aqueous solution. Four kinetic models (pseudo first-order, pseudo second-order, Elovich, and Bangham) and four isotherm models (Langmuir, Freundlich, Temkin, and Dubinin–Radushkevich) were estimated elucidate the adsorption mechanism. Current results show that the pseudo second-order model is the most appropriate kinetics for the adsorption and that the monolayer behavior (Langmuir model), rather than multilayer behavior (Freundlich equation), is more suitable to describe the adsorption. The adsorption process was confirmed to be endothermic and spontaneous via thermodynamic study. On the other hand, via Boehm’s titration we confirmed that EG@CoFe$_2$O$_4$ can contain functional groups (carboxylic, phenolic, laconic groups and basic groups) with the respective amount of 0.020, 0.044, 0.032, and 0.156 mmol/g. The EG@CoFe$_2$O$_4$ showed high maximum adsorption capacity (98.60mg/g), and good recyclability.

1. Introduction
Great industrial developments of mankind have increasingly elevated people's living needs and, at the same time, introduced a wide range of pollution issues. One of which is the discharge of organic dyes to the environment [1,2]. In printing and dyeing industries, dyes that are derived from coal-tar based hydrocarbons are usually prevalent [3,4] and thus their unabated release have raised numerous problems due to harmful effect on human health and ecosystem around the water [5-7].

Congo Red (CR) is a direct diazo dye that is toxic to many organisms and is a suspected carcinogen and mutagen [8,9]. In addition, its low degradability, high toxicity, and high stability to photodegradation have urged for development of many CR treatment methods such as catalysts, biogas, biological, advanced oxidation processes (AOPs) and physicochemical treatments [10-12]. Among them, the adsorption method has been seen an optimal method because it is cost-effective and potentially gives high-performance dye removal. This prompts for attempts aiming at manufacturing low-cost adsorbents and investigating their efficiencies for the removal of dyes from aqueous solution. Against
CR dye, notable adsorbents might include pine bark [13] or ZnFe$_2$O$_4$/SiO$_2$/Tragacanthin gum magnetic nanocomposite, both of which shown excellent adsorption capacity toward the dye [14-15]. However, these materials were sophisticatedly synthesized, and the manufacturing underwent numerous stages. With the advent of nanotechnology, researchers have explored intensively the use of carbon nanomaterials for water purification with the hope that new and successful pathways can be opened to solve current water shortage [16-18]. Among natural sources that could be utilized to manufacture adsorbents, natural graphite is a promising candidate due to its abundance and inexpensiveness and the existence of a routine that could facilely transform it into exfoliated graphite (EG) with many surface functional groups. However, EG material is difficult to recover from the aqueous solution after wastewater treatment. To resolve this, recent studies have attempted to attach ferrites nanoparticles (NPs), such as Fe$_3$O$_4$ [19], Co$_3$O$_4$ [20], CoFe$_2$O$_4$ [21] and NiFe$_2$O$_4$ [22] into graphene structure, producing composites that could be magnetically recovered.

In this study, we synthesized EG@CoFe$_2$O$_4$ by sol-gel method. Kinetic, isotherm and thermodynamic adsorption was investigated. Besides, Boehm’ titration method was also used to determine the content of functional groups onto the surface of EG@CoFe$_2$O$_4$.

2. Materials and methods

2.1. Chemicals and instruments
The precursor used in this study was natural graphite flake (GF) with the particle size of 60 mesh, gathered and selected from Yen Bai province, Vietnam. H$_2$SO$_4$ (98%), H$_2$O$_2$ (30%) and Congo red dye were purchased from Merck. The adsorbent was characterized via XRD using the D8 Advance Bruker powder diffractometer with Cu-Kα beams used as excitation sources. S4800 instrument (Japan) operating at an accelerating voltage source (15 kV) was used to capture SEM images of the sample at the magnification of 7000x. Chemical bonds and functional groups of the as-synthesized adsorbent were elucidated via infrared FT-IR spectra obtained by the Nicolet 6700 spectrophotometer. To measure CR concentration, UV-vis spectrophotometer was used at wavelength of 500 nm.

2.2. Synthesis of EG
Microwave irradiation method was used to produce EG porous material from the natural flaky graphite [22]. First, natural flaky graphite was introduced into a mixture containing H$_2$SO$_4$ (98%), H$_2$O$_2$ (30%) at a volume ratio of 100:7 and allowed to stand at room temperature for 2 hours. Afterwards, the solid was subjected to washing and then neutralization using H$_2$O and diluted NaOH solution respectively. The powder was exfoliated by microwave irradiation (750 W, 10 sec), affording the EG that would be used for subsequent studies.

2.3. Synthesis of CoFe$_2$O$_4$
Conventional polymerized complex routine was adopted to fabricate manganese-based magnetic nanoparticle of CoFe$_2$O$_4$ [26]. First, a mixture containing 93 g of citric acid, 140 mL of ethylene glycol and distilled water (2:5 by volume) was heated to 80 °C under air atmosphere. Afterwards, the mixture was added with 0.303 g of Co(NO$_3$)$_2$·6H$_2$O, followed by heating at 130°C for 2 hours. Then, the obtained polymeric resin precursor was subjected to heating in a furnace at 1000°C for 2 h and allowed to cool down at room temperature, affording the sample for further studies.

2.4. Synthesis of EG@CoFe$_2$O$_4$
The synthesis procedure of EG@CoFe$_2$O$_4$ followed a previously reported routine [26]. First, 0.7 g of Fe(NO$_3$)$_3$·9H$_2$O and 0.25 g of Co(NO$_3$)$_2$·6H$_2$O were introduced into 50 mL of H$_2$O under stirring, followed by heating at 90 °C. Afterwards, dropwise addition of 50 mL of citric acid solution (0.02 M) was carried out, followed by stirring for 60 min. Then, the solution was added with 0.8 g of EG and had pH adjusted to weakly basic (pH 8-9) with NH$_3$ solution. After 30 min, a slow addition of NH$_3$ solution
for the second time into the beaker (pH 10) was carried out. The mixture was dried at 80 °C and calcined at 700 °C during 120 min to obtain as-received sample.

2.5. Experimental batch

The batch experiments were carried out to evaluate adsorption capacity of the synthesized EG toward CR dye. First, the adsorbent was added to 100 mL of CR solution (20–60 mg/L), followed by agitation on a shaking platform. Preliminary results suggested the period after which the adsorption attained the equilibrium was 210 min. After the reaction has finished, a sample of the solution was withdrawn using a syringe and had its dye concentration determined by using UV-vis spectrophotometer at 500 nm.

Following formula was used to determine the removal efficiency (H%) and adsorption capacity (Q):

\[ H(\%) = \left(1 - \frac{C}{C_0}\right) \times 100 \]  
\[ Q_t = \left(\frac{C_0-C}{m}\right) \times V \]

where \( C_0 \) and \( C \) are, respectively, the initial and equilibrium dye concentrations, \( V \) is the volume of solution, and \( m \) represents the weight of adsorbent.

3. Results and discussion

3.1. Adsorption kinetics

Herein, the behavior of adsorption process was examined by estimating four kinetics including Pseudo-first-order, Pseudo-second order, Elovich model and Bangham model [27,28]. The results were given in table 1 and table 2. The coefficient of determination, \( R^2 \), was determined to assess the model compatibility to the data. Thus, high \( R^2 \) values show a good agreement between experimental and data predicted from the corresponding model.

From table 1, \( R^2 \) values of the pseudo second-order model (0.99983–0.99990) were so far higher than those of pseudo first-order model. The figure 1 (a, b) also confirmed the best linearity of pseudo second-order model. Similar trends could also be deduced with the CoFe\(_2\)O\(_4\) adsorbent. Specifically, table 2 and figure 2(a, b) presents \( R^2 \) (0.98934–0.99709) what were better than the other values (0.79212-0.93388). Moreover, the error between \( Q_e \) value and \( Q_2 \) value is minimal.

Thus, the pseudo second-order model is recommended as the appropriate model to describe the adsorption of CR toward EG@CoFe\(_2\)O\(_4\) and CoFe\(_2\)O\(_4\), affirming the chemisorption as the effective mechanism that controls the adsorption process. This result also accept the role of electrostatic attraction between adsorbate and adsorbent through rate-controlling steps.

On the other hand, Elovich and Bangham models are two equations can be used to determine the adsorption kinetic of CR onto EG@CoFe\(_2\)O\(_4\) and CoFe\(_2\)O\(_4\). According to the results from table 1, Bangham model seem to fit well with the experimental data since their goodness of fit coefficients (\( R^2 = 0.8469-0.96706 \)) are much higher than those by Elovich model (\( R^2 = 0.78181-0.94339 \)). From the analysis results, it was revealed that the adsorption data of CR over CoFe\(_2\)O\(_4\) suggested that Bangham model (\( R^2 = 0.94306-0.97319 \)) is more fitted than Elovich (0.92916-0.97017), which was only better fitted with the adsorption data in the CR concentration range at 30-60 mg/L. Otherwise, the CR adsorption rates (\( \alpha \), mg/g.min) far exceeded CR desorption rates (\( \beta \), g/mg) over EG@CoFe\(_2\)O\(_4\) and CoFe\(_2\)O\(_4\). Moreover, the CR adsorption rates (\( \alpha \), mg/g.min) onto EG@CoFe\(_2\)O\(_4\) were higher than onto CoFe\(_2\)O\(_4\). This indicates that the adsorption of CR towards EG@CoFe\(_2\)O\(_4\) was more favourable than over CoFe\(_2\)O\(_4\).
Figure 1. Kinetic models for adsorption of CR using EG@CoFe$_2$O$_4$ (a) Pseudo-first-order, (b) Pseudo-second-order, (c) Elovich model, and (d) Bangham model.

Figure 2. Kinetic models for adsorption of CR using CoFe$_2$O$_4$ (a) Pseudo-first-order, (b) Pseudo-second-order, (c) Elovich model, and (d) Bangham model.
Table 1. Kinetic model parameters for adsorption of CR using EG@CoFe₂O₄

| Models                  | Parameters          | Concentrations [mg/L] |
|-------------------------|---------------------|-----------------------|
|                         |                     | 20  | 30  | 40  | 50  | 60  |
| Pseudo-first-order      |                     | 0.0225 | 0.0150 | 0.0083 | 0.0143 | 0.0179 |
| (3):                    | k₁ [min⁻¹/[mg/L]¹/α] | 3.56  | 6.89  | 11.01  | 21.95  | 8.19  |
|                         | Q₁ [mg/g]           | 0.89184 | 0.88621 | 0.83756 | 0.97057 | 0.89939 |
|                         | log (Q₁, Q₁) = log (Q₁, k₁ T) R² | 0.0219 | 0.0083 | 0.0039 | 0.0020 | 0.0069 |
|                         |                     | 38.18 | 58.55 | 78.00  | 93.89  | 99.01  |
|                         |                     | 0.99999 | 0.99999 | 0.99887 | 0.99883 | 0.99992 |
| Pseudo-second-order     | k₂ [g/[mg.min]]     | 2318.55 | 8434.41 | 1619.66 | 426.77 | 2656.09 |
| (4):                    | Q₂ [mg/g]           | 0.7811 | 0.91989 | 0.93366 | 0.94339 | 0.8879 |
|                         | R²                  | 0.13635 | 0.12042 | 0.11687 | 0.08991 | 0.13139 |
|                         |                     | 0.16366 | 0.19781 | 0.18775 | 0.18398 | 0.04864 |
| Elovich (5)             | β [g/mg]            | 0.1978 | 0.9198 | 0.84 | 0.8469 | 0.88444 |
|                         | α [mg/[mg.min]]     | 0.0219 | 0.0083 | 0.0039 | 0.0020 | 0.0069 |
|                         | R²                  | 0.99999 | 0.99999 | 0.99887 | 0.99883 | 0.99992 |
| Bangham (6)             | kₐ [mL/[g/L]]       | 0.13635 | 0.12042 | 0.11687 | 0.08991 | 0.13139 |
|                         | R²                  | 0.16366 | 0.19781 | 0.18775 | 0.18398 | 0.04864 |
|                         |                     | 0.13635 | 0.12042 | 0.11687 | 0.08991 | 0.13139 |

Table 2. Kinetics model parameters for adsorption of CR using CoFe₂O₄

| Models                  | Parameters          | Concentrations [mg/L] |
|-------------------------|---------------------|-----------------------|
|                         |                     | 20  | 30  | 40  | 50  | 60  |
| Pseudo-first-order      |                     | 0.0243 | 0.0255 | 0.0195 | 0.0207 | 0.0162 |
| (3):                    | k₁ [min⁻¹/[mg/L]¹/α] | 19.42  | 31.49  | 28.13  | 29.46  | 16.59  |
|                         | Q₁ [mg/g]           | 0.93388 | 0.80165 | 0.79212 | 0.90297 | 0.89697 |
|                         | log (Q₁, Q₁) = log (Q₁, k₁ T) R² | 0.0227 | 0.0083 | 0.0039 | 0.0020 | 0.0069 |
|                         |                     | 30.08 | 40.70  | 45.70  | 48.22  | 39.51  |
|                         |                     | 0.99709 | 0.99503 | 0.98934 | 0.99527 | 0.99356 |
| Elovich (5)             | β [g/mg]            | 0.2607 | 0.1953 | 0.1930 | 0.1659 | 0.2645 |
|                         | α [mg/[mg.min]]     | 31.4832 | 41.6771 | 86.3147 | 50.9793 | 360.5440 |
|                         | R²                  | 0.97579 | 0.97017 | 0.93842 | 0.95447 | 0.92916 |
| Bangham (6)             | kₐ [mL/[g/L]]       | 0.03035 | 0.02663 | 0.02517 | 0.01893 | 0.01742 |
|                         | R²                  | 0.28722 | 0.25533 | 0.20561 | 0.21529 | 0.14132 |
|                         |                     | 0.975 | 0.97319 | 0.95719 | 0.96749 | 0.94306 |
3.2. Adsorption isotherm

The fitting of the adsorption equilibrium data into various isotherm models allows for identification of the suitable isotherm model and optimal adsorbent dosage and elaborates the mechanism by which adsorbents interact with solute [29,30]. Herein, four isotherm models: Langmuir, Freundlich, Temkin and Dubinin–Radushkevich (D–R) isotherm models were adopted and the R² values were used as the indicator to evaluate the well fitness.

The Langmuir, Freundlich, Temkin and D–R model parameters are listed in table 3. It is shown that CR adsorption onto EG@CoFe₂O₄ and CoFe₂O₄ seems to follow the Langmuir model, as evidenced by slightly higher R² compared to those of other models. This suggests that the solute might be adsorbed onto a surface with finite identical sites via monolayer adsorption. Figure 3 presents isothermal models of CR adsorption onto EG@CoFe₂O₄ and CoFe₂O₄ at varying initial CR Concentration (20, 30, 40, 50, 60 mg/L). Langmuir isotherm fittings were the most appropriate isotherm to describe the data, indicated by high linear correlation coefficient (R²) of 0.99714 and 0.967 with EG@CoFe₂O₄ and CoFe₂O₄, respectively. It is visually indicated that the isotherms data points were linear over the whole CR concentration range, demonstrating that sorption data fit well with the Langmuir model. Moreover, drawing from table 3, respective coefficients of 1/n were 0.2206, 0.1604 onto EG@CoFe₂O₄ and CoFe₂O₄, respectively, indicating a favourable adsorption.

Table 3. Parameters of Langmuir, Freundlich, Temkin and Dubinin–Radushkevich isotherms for adsorption of dyes onto EG@CoFe₂O₄ and CoFe₂O₄, respectively.

| Model        | Parameters                                                                 | EG@CoFe₂O₄ | CoFe₂O₄ |
|--------------|-----------------------------------------------------------------------------|-------------|---------|
| Langmuir (7):| \( \frac{1}{Q_r} = \left( \frac{1}{Q_m K_L} \right) \frac{1}{C_r} + \frac{1}{Q_m} \) | 2.2651     | 7.0786  |
|              | \( k_L \) [L/mg]                                                           |             |         |
|              | \( Q_m \) [mg/g]                                                           | 102.669     | 41.067  |
|              | \( R_L = \frac{1}{1+K_L C_o} \)                                           | 7.3×10⁻³    | 2.4×10⁻³ |
|              | \( R^2 \)                                                                   | 0.99714     | 0.967   |
| Freundlich (8):| \( \ln Q_r = \ln K_F + \frac{1}{n} \ln C_r \)                              | 63.36       | 25.01   |
|              | \( k_F \) [mg/g]/[mg/L]⁻¹ⁿ                                                | 0.2206      | 0.1604  |
|              | \( R^2 \)                                                                   | 0.93715     | 0.40498 |
| Temkin (9):  | \( Q_r = B_T \ln K_T + B_T \ln C_r \)                                     | 14.1751     | 5.7023  |
|              | \( k_T \) [L/mg]                                                           | 0.117       | 0.064   |
|              | \( B_T \)                                                                   | 0.93958     | 0.36522 |
| D–R (10):    | \( \ln Q_r = \ln Q_m - BE^2 \)                                            | 2.56        | 0.172   |
|              | \( Q_m \) [mg/g]                                                           | 83.867      | 43.460  |
|              | \( E \) [J/mol]                                                            | 441.94      | 170.49  |
|              | \( R^2 \)                                                                   | 0.74324     | 0.80043 |
3.3. Thermodynamic parameters

The intercept and the slope derived from plotting $\ln(K_C)$ against $1/T$ might be used to calculate values of enthalpy change ($\Delta H^o$) and entropy change ($\Delta S^o$). Gibb’s free energy $\Delta G^o$ is given by

$$\Delta G^o = -RT \ln(K_C)$$

(3)

where $R$ is the gas constant, $T$ is the absolute temperature and $K_C$ is thermodynamic equilibrium constant was calculated by $K_C = \frac{q_e}{C_e}$, where $K_c$ was calculated by

$$\ln K_C = -\left(\frac{\Delta H^o}{R} \frac{1}{T} + \frac{\Delta S^o}{R}\right).$$

(4)

Table 4 summarized free energy change and thermodynamic parameters. First, negativity of the enthalpy change indicates the exothermic nature of the CR adsorption onto EG@CoFe$_2$O$_4$. On the other hand, the adsorption process is spontaneous, reflected by the negative value of Gibb’s free energy, which is more negative with elevating temperature. Apparently, at higher temperatures, at around 40, higher adsorption has actually occurred.

| Samples | Temp [K] | $\Delta G^o$ [kJmol$^{-1}$] | $\Delta H^o$ [kJmol$^{-1}$] | $\Delta S^o$ [Jmol$^{-1}$K$^{-1}$] |
|---------|----------|-----------------|-----------------|-----------------|
| 1       | 283      | -482.5          | 57250           | 205.01          |
| 2       | 293      | -821.8          |                  |                 |
| 3       | 303      | -1161.1         |                  |                 |
| 4       | 313      | -1500.4         |                  |                 |
3.4. Boehm’ titration method

The Boehm assay was used to differentiate and quantify functional groups, in which different bases including NaHCO$_3$, Na$_2$CO$_3$ and NaOH were used. To be specific, NaHCO$_3$ ($pK_{NaHCO_3} = 6.37$) neutralizes only carboxyl groups on the carbon surface, Na$_2$CO$_3$ ($pK_{Na_2CO_3} = 10.25$) titrates carboxyls and lactones, and NaOH ($pK_{NaOH} = 15.74$) neutralizes carboxyl, lactone and phenolic groups [17]. In this report, quantity of functional groups was expressed as mmol of Phenolic, lactic, carboxylic and basic per gram of EG@CoFe$_2$O$_4$. The results are shown in table 5. It is shown that EG@CoFe$_2$O$_4$ contained functional groups (carboxylic, phenolic, lactic groups and basic groups) with the amount of 0.020, 0.044, 0.032, and 0.156 mmol/g, respectively. Unlike EG@CoFe$_2$O$_4$, these functional groups were not found for CoFe$_2$O$_4$. Thus, it is suggested that EG@CoFe$_2$O$_4$ might exhibit improved adsorption of CR compared with CoFe$_2$O$_4$ where no surface functional groups were found.

| Sample     | Carboxylic | Phenolic | Lactic  | Number of oxygenated functional groups [mmol/g] | Total basic [mmol/g] |
|------------|------------|----------|---------|-----------------------------------------------|---------------------|
| CoFe$_2$O$_4$ | 0          | 0        | 0       | 0                                              | 0                   |
| EG@CoFe$_2$O$_4$ | 0.020      | 0.044    | 0.032   | 0.096                                          | 0.156               |

3.5. Recyclability evaluation of EG@CoFe$_2$O$_4$

Reutilization is a major advantage when using an adsorbent in water treatment processes. Recyclability assessment also provides useful information about adsorption stability during the adsorbent cycles. The recycling capability of the EG@CoFe$_2$O$_4$ was quickly tested and the results were shown in figure 6. After every run, appreciable reduction in adsorption capability was observed. In detail, the degradation ratio of CR by EG@CoFe$_2$O$_4$ adsorbent reached almost 100% after the first cycle, then quickly decreased in following runs and ultimately only attained 12.2 % after the fifth run. This is possibly explained to repeated exposure of active sites to CR molecules that resulted in reduced adsorption capacity.
4. Conclusion
In this study, we demonstrated that the adsorption of CR at different CR concentrations over EG@CoFe$_2$O$_4$ followed chemisorption mechanism (pseudo-second order), and monolayer adsorption behavior (Langmuir equation). The adsorption process was confirmed to be endothermic and spontaneous in nature. Compared with CoFe$_2$O$_4$, EG@CoFe$_2$O$_4$ showed remarkably higher maximum adsorption capacity, which was confirmed by results obtained from Langmuir model. These results are consistent with the results of the Boehm’s titration and suggest the promising potential of EG@CoFe$_2$O$_4$ in adsorption of CR dye from aqueous solution.

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