



## Highly efficient Co<sub>3</sub>O<sub>4</sub>/GO adsorbent recycled from spent Lithium-Ion

### Batteries toward cationic dye from aqueous solutions



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#### Abstract

In this study, the hydrothermal approach was used to extract cobalt oxide (Co<sub>3</sub>O<sub>4</sub>) nanoparticles and graphene oxide (GO), respectively from the cathode and the anode of spent lithium-ion batteries (LiBs). To gather the benefits of both Co<sub>3</sub>O<sub>4</sub> and GO, the Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite was prepared via hydrothermal method. The prepared materials have been characterized by Fourier transform infrared (FT-IR), X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and nitrogen adsorption-desorption techniques. The characteristic analysis confirms the successful preparation of Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite with a BET surface area of 40.42 m<sup>2</sup>/g. Due to high porous structure, and the presence of GO layers, the prepared Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite displayed high adsorption capacity toward methylene blue (MB) from the aqueous solution. Various adsorption parameters affecting the overall adsorption efficiency such as pH, adsorbent doses, contact time, and initial MB dye concentrations have been systematically investigated. The highest removal efficiency of MB dye onto the Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO was achieved at alkaline conditions (pH ≈ 8.5) with a removal efficiency of 94% and 99%, respectively. The kinetic analysis confirmed that the adsorption of MB dye onto Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO adsorbents fitted with the pseudo-second order model. The isotherm results indicated that the adsorption of MB dye Co<sub>3</sub>O<sub>4</sub> and the Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite obeyed the Langmuir isotherm model, and the estimated maximum adsorption capacity of the Co<sub>3</sub>O<sub>4</sub> and the Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite are 64.94 and 68.97 mg/g, respectively. This study opens a way for converting waste LiBs components into valuable nanomaterials to be applied for water treatment applications.

**Keywords:** Recycling; metal oxides; lithium-ion batteries (LiBs); Graphene oxide; hydrothermal method; organic pollutants.

#### 1. Introduction

In modern society, lithium-ion batteries (LiBs) are the most attractive power source for portable electronic products such as cell phones and laptops. LiBs consist of a cathode, anode, separator, and organic electrolyte [1]. A combination of cathode active components (LiCoO<sub>2</sub>), a binder (polyvinylidene fluoride) (PVDF), an electrical conductor, and other additives make up the cathode electrode, which is made of an aluminium plate. The anode electrode is a copper plate covered with a mixture of carbon graphite, an electrical conductor, some additives, and a PVDF binder [1]. Numerous devices, including mobile phones, digital cameras, some electric cars, and others use LiBs [1]. The LiBs may be able to withstand temperatures of up to 3000 °F. The productivity of LiBs is quite high, as it was 2.05 billion in 2005 rose to 5.86 billion in 2012 globally, and increased to 7.84 billion in 2016 and is still rising [2]. The metal content of spent LiBs includes cobalt, aluminium, lead, and lithium, as well as, the presence of hazardous make them unsafe [3]. Some components of spent LiBs must be recovered or extracted to preserve the environment as well as resources [4,5]. This is because they are difficult to decompose or break down and may cause soil and underground water pollution and release some toxic gases that could affect human health. Cobalt oxide is a main component in cathode of LiBs [1,6]. Usually, metals can be extracted from waste batteries or other sources using a variety of techniques, including hydrometallurgical [7], and pyrometallurgical [8]. In the pyrometallurgical process, used batteries are selectively volatilized at high temperatures, then condensed. It is commonly used in the battery recycling process due to its simplicity; however, some toxic gases are emitted. However, the hydrometallurgical process, which involves

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dissolving battery electrodes in strong acids and then recovering the metal by precipitation, extraction, electrodeposition, or the hydrothermal technique, provides a less harmful way for recovering metal from wasted batteries [9].

Currently, water pollution is considered a serious problem that needs to be taken into consideration to keep our water clean and safe.[10,11] From various pollutants dyes are considered the most common source of water pollution in textile, cosmetics, leather, food, pharmaceutical, paint, varnish, pulp, and paper industries. These dyes harm the aquatic ecosystem by lowering the aesthetic value of water features and obstructing light penetration [12]. Some dyes have toxic, mutagenic properties, and can even cause cancer when combined with their metabolites. These dyes also cause problems for human renal, liver, and nervous system dysfunction. They are difficult to decolorize due to their complicated structure [13,14]. Thus, there is great interest from environmentalists and researchers to remove these harmful compounds from wastewater [15-19]. Additionally, dyes are thought to be a contributing factor in eutrophication and pollution. There are various methods for treating water-containing dyes including chemical oxidation, adsorption [20,21], and photodegradation [16,17]. Adsorption is the most efficient and cost-effective option for dye recovery from water [22-24]. Because adsorption procedures are affordable, highly successful, and economically viable, they are the most often used of all these approaches [23,25]. The adsorbent materials and their morphologies, which are essential to the total adsorption performance, are the primary problem facing the adsorption process [26,27]. In the last decades, a lot of materials have been used as effective adsorbent materials such as carbons [23,28,29], MOFs [30], metal oxides [31,32], two-dimensional materials,[33] agriculture biowaste materials, [34,35] waste-derived adsorbents,[36].....etc.

Here, Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite has been prepared from the waste LiBs using a hydrothermal technique to be utilized as adsorbent for MB cationic dye from aqueous solution. The Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite displayed higher adsorption efficiency (R=99%) than that achieved by pure Co<sub>3</sub>O<sub>4</sub> (R=94%). Such higher efficiency is mainly assigned to the presence of GO layers. The isotherm analysis confirms that the adsorption of MB onto the Co<sub>3</sub>O<sub>4</sub> and the Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite obeyed Langmuir isotherm model which indicates a chemical adsorption process. The estimated maximum adsorption capacity of monolayers MB dye onto the Co<sub>3</sub>O<sub>4</sub> and the Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite are 64.94 and 68.97 mg/g, respectively. This study suggests the potential application of Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite as effective adsorbent for wastewater treatment.

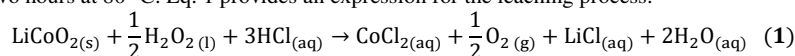
## 2. Experimental (Materials and Methods)

### 2.1 Chemicals and reagents

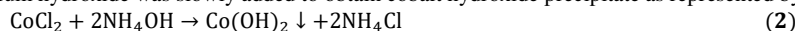
All chemicals used in this study were analytical grade. Sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), phosphoric acid (H<sub>3</sub>PO<sub>4</sub>), ammonium hydroxide (NH<sub>4</sub>OH), potassium permanganate (KMnO<sub>4</sub>), sodium nitrate (NaNO<sub>3</sub>, 99%), sodium hydroxide (NaOH, ≥ 98%, pellets anhydrous), hydrochloric acid (HCl, 37%), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), absolute ethanol (C<sub>2</sub>H<sub>6</sub>O, ≥ 95%), and methylene blue (MB; C<sub>16</sub>H<sub>18</sub>ClN<sub>3</sub>S, ≥ 95%) were purchased from Merck, Darmstadt, Germany. All preparations in our study have been performed with distilled water obtained from an ultra-pure purifier (Ulupure, resistivity ≥ 18.2 MΩ).

### 2.2 Extraction Co<sub>3</sub>O<sub>4</sub> from the cathode

Samsung Dead 3.8 V LiBs underwent physical disassembly and separation. After washing the cathode powder in warm water for two hours at 3000 rpm using a magnetic stirrer to get rid of any contaminants, it was dried for twelve hours at 80 °C. After dissolving 2 g of dry powder in 80 mL of 3 M HCl and 5 mL of H<sub>2</sub>O<sub>2</sub> as a leaching solution, the mixture was stirred magnetically for two hours at 80 °C. Eq. 1 provides an expression for the leaching process.



After that, ammonium hydroxide was slowly added to obtain cobalt hydroxide precipitate as represented by Eq. 2.



After that, the autoclave was placed in an oven set at 150 °C for 4 h, and the suspension solution from Eq. 2 was added. After being cleaned with distilled water and ethanol, the precipitate was dried for 24 h at 100 °C. To prepare cobalt oxide, the dried sample was calcined for 2 h at 480 °C.

### 2.3 Preparation of GO from the anode

First, the contaminants have been eliminated, and then the carbon powder from the anode of LiBs was separated and washed with hot distilled water and dried at 100 °C for 12 h. based on the modified Hummer approach [37]. Then, 3 mL of concentrated H<sub>3</sub>PO<sub>4</sub> and 27 mL of concentrated H<sub>2</sub>SO<sub>4</sub> (V/V ratio 9:1) were combined and agitated for 15 minutes. Next, using a magnetic stirrer, 0.225 g of drying powder was added to the acid mixture mentioned above. After that, 1.32 g of KMnO<sub>4</sub> was gradually added to the solution while being guided for 24 h, or until the colour turned dark green. The hue changed to bright green when precisely 0.675 mL of H<sub>2</sub>O<sub>2</sub> solution was gradually added to the aforesaid solution in an ice bath while being stirred magnetically. After being cleaned with 10 mL of 30% HCl and distilled water, the resulting powder was dried for 24 h at 100 °C.

### 2.4 Preparation of Co<sub>3</sub>O<sub>4</sub>/GO composite:

Exactly 0.3 g of GO powder was mixed with 70 mL of CoCl<sub>2</sub> solution, and the mixture was stirred magnetically for 30 minutes. After that, NH<sub>4</sub>OH was gradually added under magnetic stirrer until the Co(OH)<sub>2</sub> was fully prepared. Then, the

mixture was placed in an autoclave and heated to 160 °C for 4 h. The resulting precipitate was then washed with distilled water, dried at 100 °C for 24 h, and then burnt at 480 °C for 2 h. The obtained material is called Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite.

## 2.5 Characterization and measurements

Several techniques were used to characterize the prepared GO and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite derived from waste LiBs. FT-IR used to determine the functional groups, and the FT-IR spectrum was obtained in the 4000-400 cm<sup>-1</sup> regions at a resolution of 2 cm<sup>-1</sup> on a Nicolet spectrophotometer. The crystal structure was investigated by XRD on a monochromatic Cu-K<sub>α</sub> radiation source running at 40 kV. The surface morphology was examined by SEM (JSM 7100F FESEM; Zeiss Ultra Plus). TEM images were obtained by a JEM-2100F field emission microscope (JEOL Ltd., Japan). The surface area was investigated by nitrogen adsorption-desorption on Micromeritics ASAP 2020 HD88 system. The surface area and the pore size distribution were determined by Brunauer-Emmett-Teller (BET) and Barrett-Joyner-Halenda (BJH) models, respectively. The point of zero charge (pHPZC) was determined using the drift method.[38] The experiments were conducted by 20 mL of 0.1 M NaCl solutions with initial pH (pH<sub>i</sub>) from 2 to 12 adjusted by 0.1M HCl and 0.1M NaOH solutions. To each solution, 0.05 g of Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite were added and kept under shaking for 24 h, and then the final pH values (pH<sub>f</sub>) of the supernatant liquid were measured. By plotting the pH<sub>i</sub> vs. the pH<sub>f</sub>, the pH values of the pHPZC were determined. [44]

## 2.6 Adsorption MB dye

MB dye adsorption was done in the dark in a 50 mL flask using a magnetic stirrer. The adsorption efficiency of the produced GO and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite was examined using the batch adsorption technique, considering the influences of solution pH, contact duration, dye concentration, and adsorbent dosage. At a starting concentration of 50 mg/L, 0.02 g of adsorbent material was combined with 25 mL of dye solution in each experiment. Solutions of 0.1 M HCl and 0.1 M NaOH were used to change the pH of the mixture. A UV-visible spectrophotometer set to 664 nm was used to determine the residual concentration of the MB dye solutions. The removal percent (R) was calculated using Eq. 3.

$$R (\%) = \frac{C_i - C_f}{C_i} \times 100 \quad (3)$$

Where, C<sub>i</sub> and C<sub>f</sub> are the concentrations of MB in initial and final solutions, respectively. The amount of MB adsorbed q (mg/g) was calculated from the mass balance equation as given by Eq. 4 .

$$q = \frac{(C_i - C_f)}{W} V \quad (4)$$

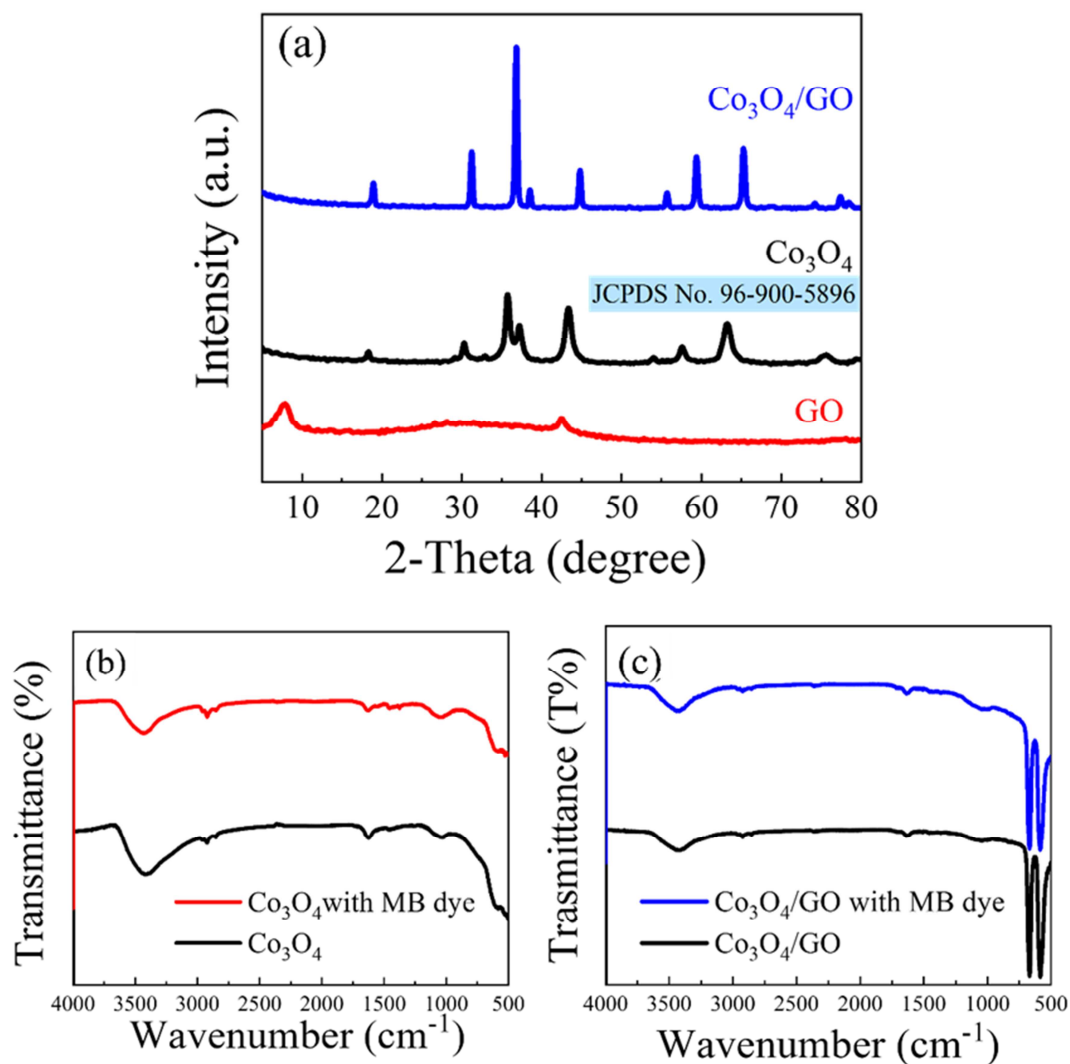
Where W (g) is the mass of adsorbent, and V is the dye volume (L).

## 3. Results and discussion

### 3.1 Adsorbent characterization

As in Fig. 1, the XRD of the Co<sub>3</sub>O<sub>4</sub> after thermal annealing at 480 °C for 2 h displayed that all diffraction peaks indexed into the face-centered cubic Co<sub>3</sub>O<sub>4</sub> structures (spacing group Fd3m) (JCPDS No. 96-900-5896) in positions 2θ of 19.04°, 30.34°, 36.93°, 37.64°, 44.92°, 54.79°, 57.51° and, 63.40°. These peaks were associated with the (111), (220), (311), (222), (400), (422), (511), and (440) planes. No traces of other phases or impurities, which undisputedly indicate the formation of single crystalline Co<sub>3</sub>O<sub>4</sub>. This indicates the successful preparation of cobalt oxide. The broad peak that characterizes GO at 2θ = 9.7° and 41.2° is attributed to (002) and (001) diffraction planes respectively. This indicates successful preparation of GO. From the XRD pattern of Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite, all diffraction peaks of Co<sub>3</sub>O<sub>4</sub> and GO were observed which indicates the successful preparation of Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite.

The FTIR spectra of the Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite with and without adsorbed dyes are displayed in Fig. 1(b,c). From the FT-IR spectrum, the band at 3417 cm<sup>-1</sup> for the O-H groups because of the vibration of water-adsorbed molecules, the peak of Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite is stronger than the peak of Co<sub>3</sub>O<sub>4</sub>. Furthermore, both materials are visible at a peak that belongs to the C=C groups and it is situated at 1623 cm<sup>-1</sup>. The faint band at 582 cm<sup>-1</sup> in the spectrum may also be caused by the existence of the C-O group in the sample. Strong absorption bands can be seen at 560 and 660 cm<sup>-1</sup>, due to the vibrations of Co-O stretching in Co<sub>3</sub>O<sub>4</sub>. [39] The FTIR spectra shows that both materials are loaded with MB dye. Almost all the bands on the material surface are still visible after the adsorption of MB; however, chemisorption is indicated by the O-H band at 3417 cm<sup>-1</sup>, which is stronger than those before dye adsorption.



**Fig. 1:** (a) XRD patterns of  $\text{Co}_3\text{O}_4$ , GO, and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite from waste LiBs. FT-IR spectra of (b)  $\text{Co}_3\text{O}_4$  and (c)  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite with/without adsorbed MB dye.

TEM images of the  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite in Fig. 2 (a-c) displayed successful preparation of the  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite, in which the dark particles represent the  $\text{Co}_3\text{O}_4$  particles and the transparent layers for the GO layers. The estimated average particle size of  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite was found to be in the range from 13.1 to 41.9 nm. On the other hand, SEM analysis was performed to determine the shape and morphology of  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite under various magnifications and results were depicted in Fig. 2 (d-f). SEM images of the  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite showed that the  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite displayed a morphos structure in which the  $\text{Co}_3\text{O}_4$  particles are loaded into GO layers. This also confirms the successful preparation of  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite.

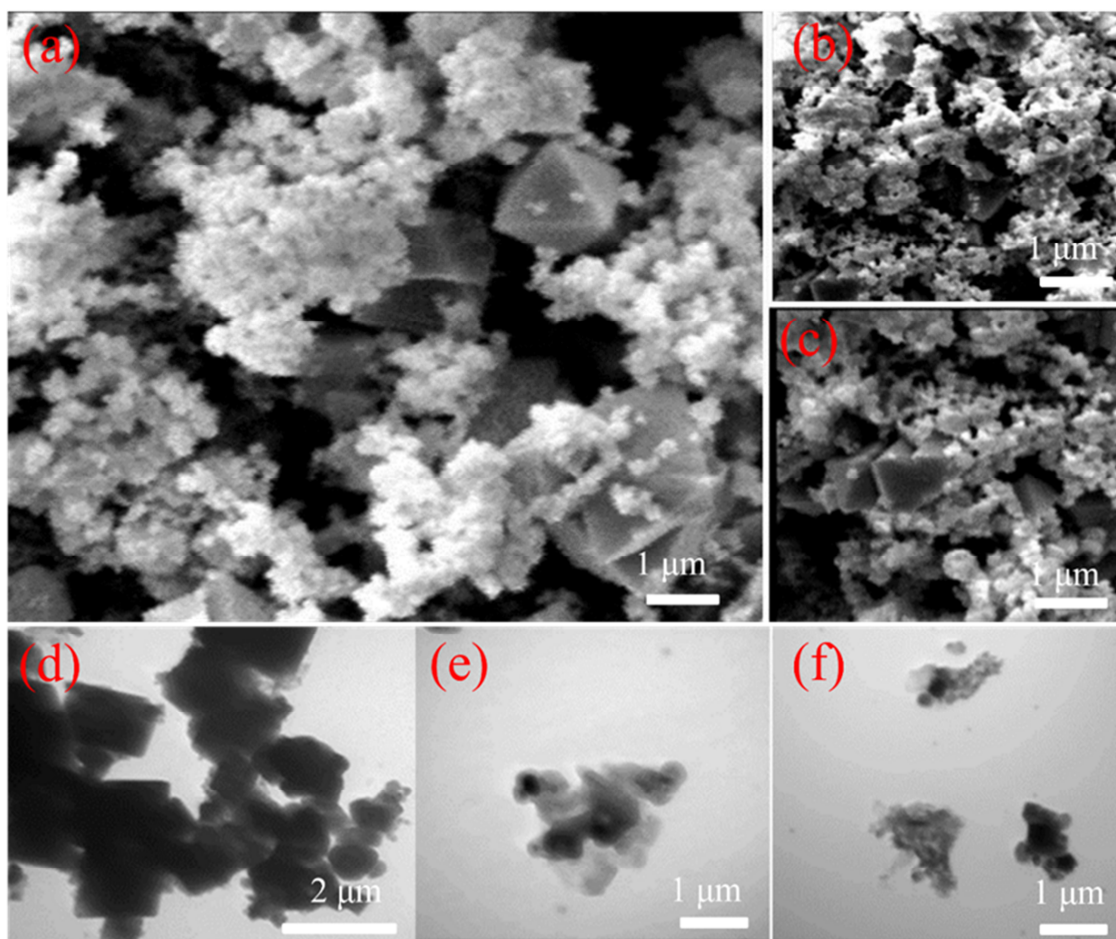


Fig. 2: SEM (a-c) and TEM (d-f) images of  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite.

As presented in **Fig. 3a**, the nitrogen adsorption-desorption isotherm of the  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite displayed type-I, which is characteristic of mesoporous materials as stated by the IUPAC categorization.[40] Additionally, the pore size distribution curve in **Fig. 3b** showed that mesopore concentrations are concentrated. The  $S_{\text{BET}}$  and the pore volume of the  $\text{Co}_3\text{O}_4/\text{GO}$  are  $40.42 \text{ m}^2/\text{g}$ , and the mean pore diameter is  $8.61 \text{ nm}$ .

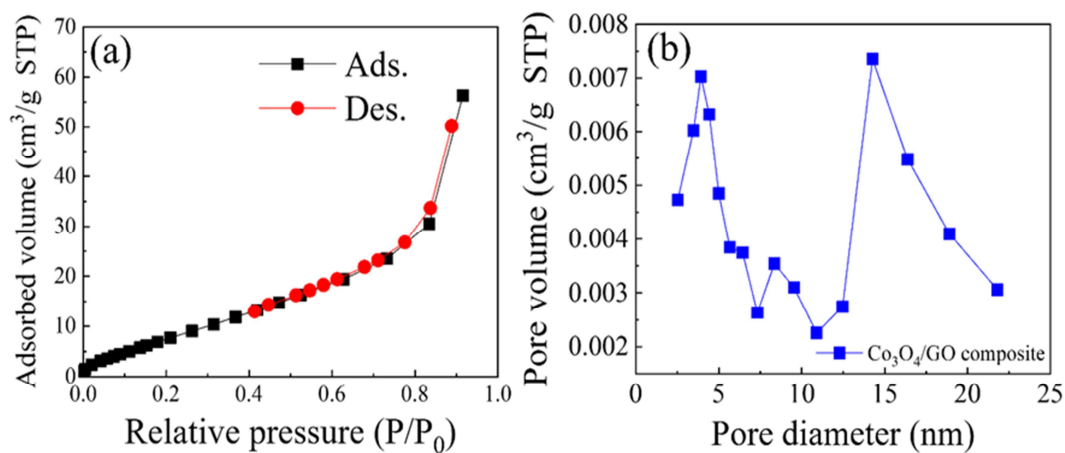


Fig. 3: (a) Nitrogen adsorption-desorption isotherms and (b) pore size distribution of the  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite.

The  $\text{pH}_{\text{PZC}}$  value is crucial for figuring out the adsorbent's surface charge, which is often influenced by the pH. Thus, throughout the adsorption process,  $\text{pH}_{\text{PZC}}$  is utilized to illustrate the forces of attraction and repulsion between the adsorbent and adsorbate. Here, we use the drift approach to calculate the  $\text{pH}_{\text{PZC}}$  based on earlier research.[32] Fig. 4 illustrates how the existence of positive surface charges is shown by the final pH rising as the system's initial pH rises. The  $\text{pH}_{\text{PZC}}$  can be determined by the point of intersection of the final pH line and the initial pH line. The estimated  $\text{pH}_{\text{PZC}}$  values are 7.55 and 7.90 for the  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite, respectively. At pH values below  $\text{pH}_{\text{PZC}}$ , the surfaces of the  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite were positive, while at pH values above  $\text{pH}_{\text{PZC}}$ , the surfaces were negative.

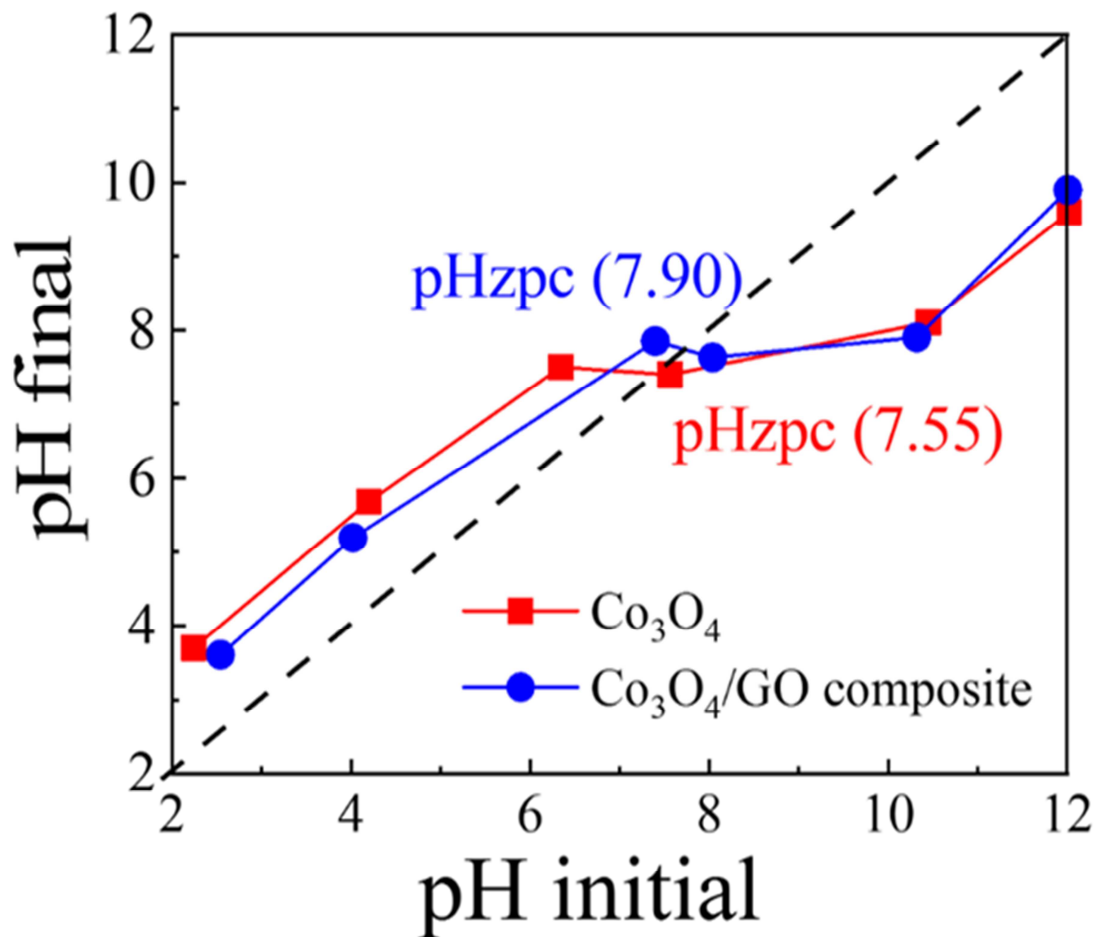


Fig. 4: (a) The  $\text{pH}_{\text{PZC}}$  of the  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite.

### 3.2 Adsorption study

#### 3.2.1 Effect of pH

The effect of pH in Fig. 5a shows the removal percentage of MB dye onto  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite remarkably varies when pH increased from 4 to 11, and the highest removal efficiency was achieved at pH 8.5. This suggests that most of the dyes were eliminated by both materials at basic conditions. For  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite, the clearance percentages of MB at pH 8.5 were 88% and 97.3%, respectively.

#### 3.2.2 Effect of contact time

The impact of the adsorption duration on the extracted materials' adsorption capability was investigated at a pH of 8.5. The dye solutions were stirred at 300 rpm with 0.02 g of adsorbent material for various time from 15 to 240 minutes. As showed in Fig. 5b, when the contact duration was extended from 15 to 240 minutes, the elimination percentage of MB rose from 17 to 94% for  $\text{Co}_3\text{O}_4$  and from 44 to 99.8% for  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite.



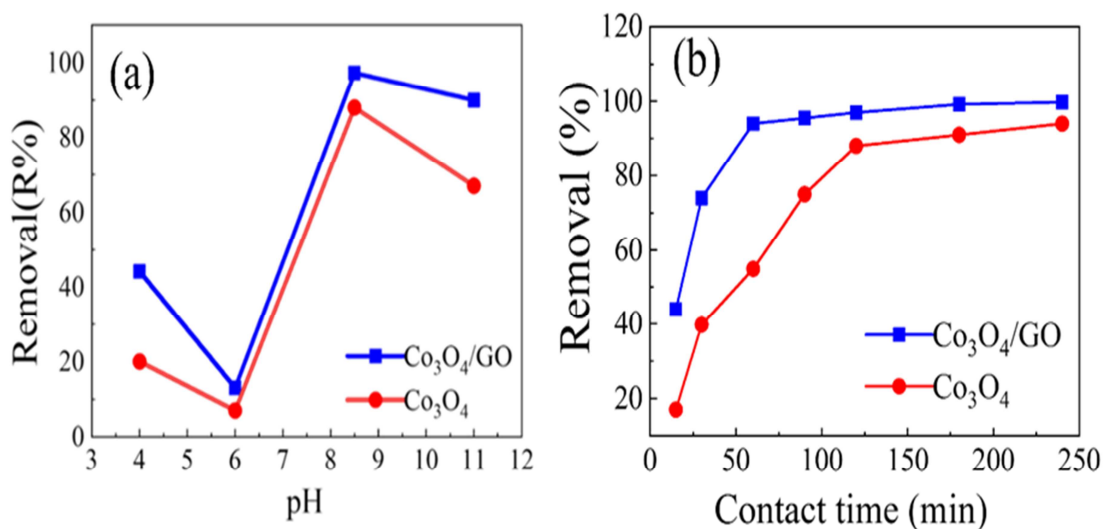


Fig. 5: Effect of (a) solution pH on the removal efficiency of MB dye from aqueous solution [ $M = 0.02$  g,  $V = 25$  mL,  $C_0 = 10$  mg/L,  $t = 1$  h,  $\text{pH} = 4-11$ ] and (b) contact time on the removal efficiency of MB dye. [ $M = 0.02$  g,  $V = 25$  mL,  $C_0 = 10$  mg L $^{-1}$ ,  $t = 15-240$  min,  $\text{pH} = 8.5$ ].

### 3.2.3 Effect of adsorbent dose

After the equilibrium phase is reached, material waste must be avoided by examining the adsorbent dose.[41] Fig. 6a displays the results of an experiment that examined the effects of different adsorbent dosages on the dye removal process from aqueous solutions. The adsorption of the MB dye at 10 mg/L is investigated using several adsorbent amounts ranging from 0.002 to 0.02 g at a constant pH of 8.5 and a contact period of 240 minutes. The removal percentage of  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite for MB dye rose gradually from 38 to 94% and from 86 to 99.80% when the adsorbent dosage was raised from 0.002 to 0.02 g. Accordingly, a larger dose implies greater surface area or active sites which increasing the overall removal efficacy.

### 3.2.4 Effect of concentration

Fig. 6b illustrates the adsorption capacities of the  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite at various MB concentrations. By raising the initial dye concentration, adsorption MB capabilities are somewhat increased. The  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  composite adsorption capabilities rose from 3.13 to 45.64 mg/g and from 3.29 to 49.6 mg/g, respectively, when the concentration increased from 10 to 200 mg/L.

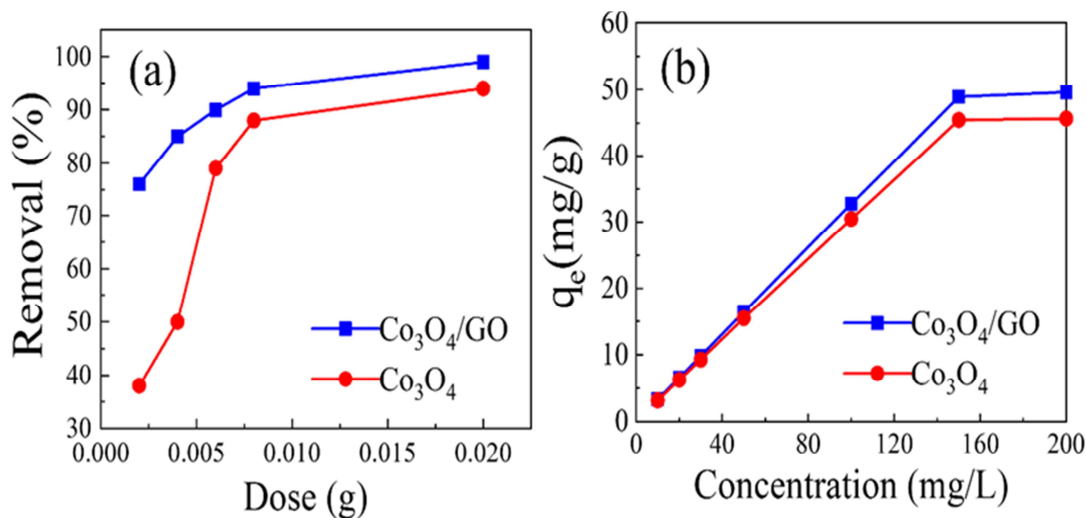


Fig. 6: Effect of (a) dose on MB removal. [ $M = 0.002-0.02$  g,  $V = 25$  mL,  $C_0 = 10$  mg/L,  $t = 240$  min,  $\text{pH} = 8.5$ ] and (b) initial MB dye concentration. [ $M = 0.02$  g,  $V = 25$  mL,  $C_0 = 10-200$  mg/L,  $t = 240$  min,  $\text{pH} = 8.5$ ].

### 3.2.5 Adsorption kinetics

To examine the kinetic behaviour of MB dye adsorption by  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite, the pseudo-first order and pseudo-second order models were employed to assess the experimental results. The pseudo-second-order (Eq. 6) and pseudo-first-order (Eq. 5) kinetic models.[42],[43,44]

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (5)$$

$$\frac{t}{q_t} = \frac{1}{q_e^2 k_2} + \frac{t}{q_e} \quad (6)$$

Where,  $q_e$  and  $q_t$  are the concentrations of MB in the solution (mg/g) at equilibrium and time  $t$ , respectively.  $k_1$  ( $\text{min}^{-1}$ ), the pseudo-first-order adsorption rate constant,  $k_2$  (g/mg. min) the pseudo-second-order adsorption rate constant.

The fitted data are plots in Fig.7, and the related kinetic parameters and their correlation coefficients are listed in Table 1. It can be observed that the experimental data of MB adsorption onto the  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite are fitted by the pseudo-second order kinetic model, as indicated by the higher correlation coefficient ( $R^2$ ) values. The estimated  $q_e$  for MB utilizing  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite were 60 and 70 mg/g, respectively. Furthermore, from the fitted data in Fig.7 and parameters in Table 1, despite the higher  $R^2$  values for the pseudo-second order models, the pseudo-first order models are also displayed higher values ( $> 0.9$ ), which indicates the presence of both physical and chemical adsorption processes. However, the higher  $R^2$  values demonstrated that the adsorption process mainly followed the pseudo-second-order model.

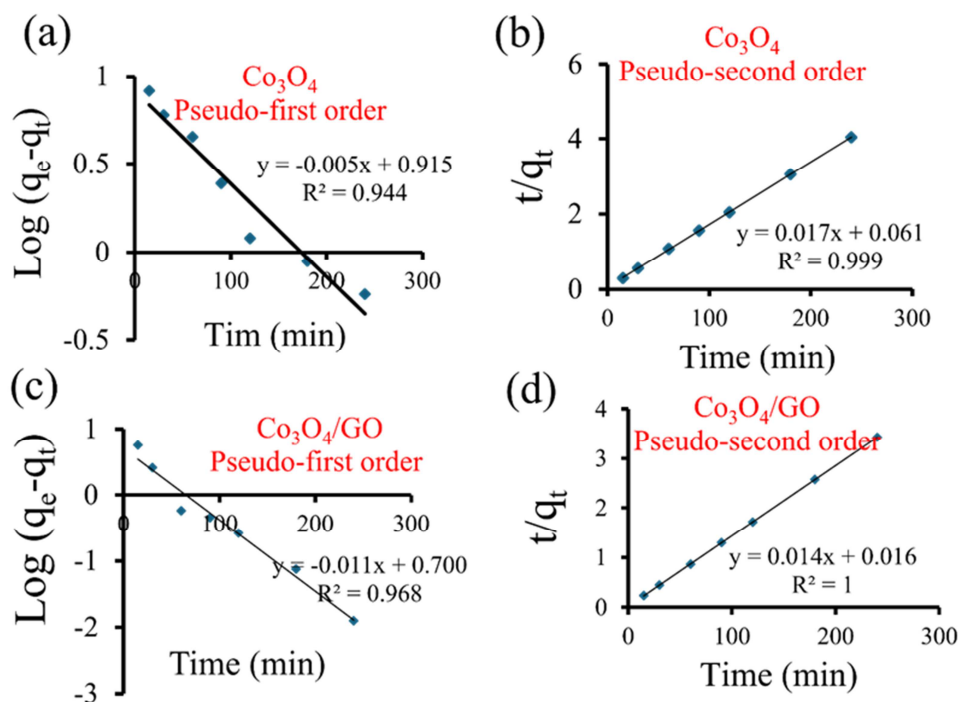


Fig. 7: Pseudo-first order and pseudo-second-order models for the adsorption of MB onto  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{O}_4/\text{GO}$  nanocomposite.

Table 1: Parameters of the pseudo-first, pseudo-second-order kinetic models.

Kinetic model	Parameters	$\text{Co}_3\text{O}_4$	$\text{Co}_3\text{O}_4/\text{GO}$ nanocomposite
Pseudo-first order	$R^2$	0.944	0.968
	$q_e$ (mg/g)	8.226	5.014
	$k_1$ ( $\text{min}^{-1}$ )	-0.012	-0.025
Pseudo-second order	$R^2$	0.999	1.0
	$q_e$ (mg/g)	60.241	70.422
	$k_2$ (g/mg.min)	0.004	0.012



### 3.2.6 Adsorption isotherms

The equilibrium states for dye adsorption on solid surfaces have been described by several mathematical models. The Freundlich and Langmuir models are often used to explain the experimental findings. The Langmuir isotherm model is used for monolayer adsorption on adsorbent surface with a limited number of active sites [43,45,46]. Langmuir model assumes that adsorption occurs on a homogenous surface and that there is no contact between the adsorbates in the surface [47,48]. Eq. 7 represents the Langmuir isotherm.

$$\frac{1}{q_e} = \left(\frac{1}{q_{\max}k_L}\right) \frac{1}{C_e} + \frac{1}{q_{\max}} \quad (7)$$

Where,  $k_L$  is a Langmuir constant associated with the energy of adsorption (L/mg)

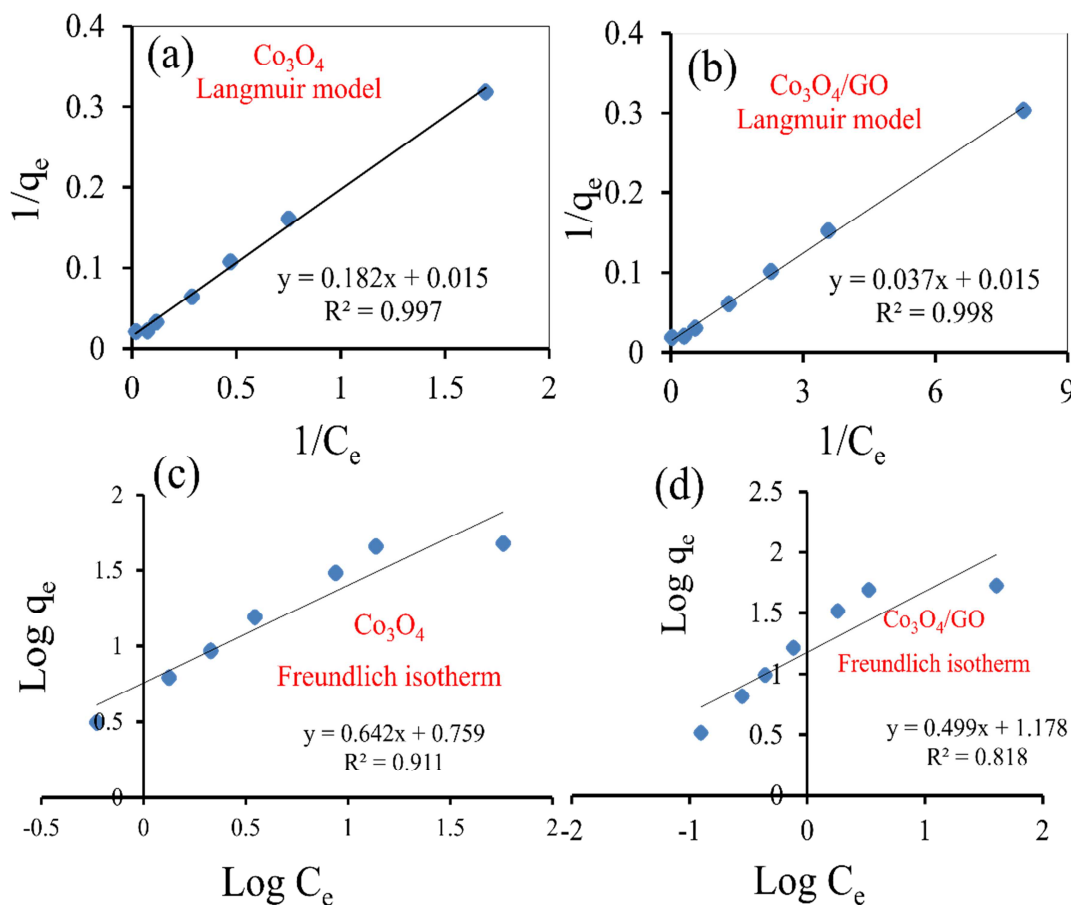
and the affinity of the binding sites, and  $q_{\max}$  (mg/g) is the maximum adsorption capacity.

The Freundlich adsorption isotherm is an equation for a heterogeneous system in which multilayer adsorption may occur on the adsorbent surface [47]. Eq. 8 provides the relationship between equilibrium liquid and solid phase capacity.

$$\log q_e = \log k_f + \frac{1}{n} \log C_e \quad (8)$$

Where,  $k_f$  and  $n$  stand for the Freundlich constants. Adsorption process favorability is indicated by the dimensionless constant  $n$ ; the adsorption capacity is indicated by  $k_f$ .

As shown in **Fig. 8** and **Table 2**, the Langmuir and Freundlich isotherms can suit the experimental data of MB adsorption onto Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite. The Langmuir isotherm model appears to better explain the experimental results based on higher  $R^2$ . **Table 2** illustrates that the  $K_L$  values for MB adsorption by Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite are higher than those for adsorption by Co<sub>3</sub>O<sub>4</sub>. This indicates that Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite and dye molecule connections are more stable than Co<sub>3</sub>O<sub>4</sub> interactions. For MB onto the Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite, the calculated  $K_L$  values are 0.085 and 0.396 L/mg, respectively.



**Fig. 8:** Langmuir and Freundlich isotherms for adsorption of MB on Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite.

**Table 2:** The parameters of Langmuir, and Freundlich isotherms for adsorption of MB onto Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite

Isotherm model	Parameters	Co <sub>3</sub> O <sub>4</sub>	Co <sub>3</sub> O <sub>4</sub> /GO nanocomposite
Langmuir	R <sup>2</sup>	0.997	0.998
	q <sub>max</sub> (mg/g)	64.935	68.966
	K <sub>L</sub> (L/mg)	0.085	0.396
Freundlich	R <sup>2</sup>	0.911	0.8176
	N	1.558	2.002
	K <sub>f</sub> (L/g)	5.742	15.056

### Conclusions

In this study, Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite has been reported from waste LiBs via hydrothermal technique for removal of MB from aqueous solution. The results revealed that the adsorption of MB onto Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite is strongly controlled by pH, contact time, dye concentration, and adsorbent weight. The maximum adsorption efficiency was achieved at pH of 8.5, and the Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite showed excellent MB adsorption with a removal efficiency of 94% and 99%, respectively. The adsorption equilibrium of MB onto Co<sub>3</sub>O<sub>4</sub> and Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite was fitted with Langmuir isotherm, as well as the pseudo-second-order kinetic model confirm the chemical adsorption process. The estimated maximum adsorption capacity of the Co<sub>3</sub>O<sub>4</sub> and the Co<sub>3</sub>O<sub>4</sub>/GO nanocomposite are 64.94 mg/g and 68.97 mg/g, respectively. The findings confirmed potential ability to utilize the prepared materials for removal of cationic dyes from aqueous solutions.

**Conflict of interest:** The authors declare there are no conflicts.

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