

## Development of a Nanoscale (Mg/Al)-LDH Coated Waterworks Sludge Sorbent for Efficient Congo Red Dye Removal – Batch Kinetics and Isotherm Studies

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### ABSTRACT

The study's aim was to develop a novel sorbent for removing the color Congo red (CR) from water by precipitating (Mg/Al)-layered double hydroxide (LDH) at the nanoscale onto the waterworks sludge surface as a byproduct. To achieve an effective sorbent with the removal of over 91.19%, The utilization of nanoparticles, the addition of 1 g of sludge to 50 mL of water, and a molar ratio of 2 (Mg/Al) were the optimal production conditions for sorbent. In batch experiments, the optimal operating parameters were found to be 0.5 g of adsorbent in 50 mL of CR, pH of CR solution equal to 3, and a contact time of 3 hours at 200 rpm with 25 mg/L dyes. According to the study, the adsorption capacity was 23.576 mg/g. Additionally, the pseudo-second-order and Langmuir models provided accurate descriptions of the sorption data.

**Keywords:** layered double hydroxides, Langmuir model, coating, simulation and sorption.

### INTRODUCTION

The present-day surface and subsurface water resources face significant environmental threats from dyes, as they can significantly reduce the dissolved oxygen content in water, posing a hazardous situation for aquatic life. Furthermore, the breakdown products of these substances can be carcinogenic and endanger the survival of aquatic organisms (Thabede et al., 2020; Waleed Khalid and Salman, 2019). Dyes can cause various harmful effects such as disrupting wastewater clearance, inducing ulcers on the skin and mucous membranes, causing nausea, hemorrhage, dermatitis, and increasing skin sensitivity (Gupta et al., 1990; Hummadi, 2021). Businesses involved in paper production, inkjet printing, food preparation, cosmetics manufacturing, and leather dyeing often generate industrial effluent that contains pigments. Acid dyes, including Acid Blue, Methyl Orange, and Orange (I, II), are utilized in various industries for their versatile properties (Faisal et al., 2022; Finish

and Naife, 2021; Jawad and Naife, 2022; Salleh et al., 2011; Wang et al., 2019).

An anionic dye with acidic qualities, Congo Red (CR), is frequently used in aquaculture and animal husbandry, especially in commercial fish hatcheries, as an antifungal agent (Frid et al., 2007). However, due to its negative health effects, such as amyloidosis, the use of this dye has been prohibited in many countries worldwide, including Europe and the United States. Congo Red dyes must be separated from polluted aqueous solutions. Several methods, including adsorption, coagulation-flocculation, ion exchange, mineralization, photocatalysis, and ultrasonic irradiation, have been proposed to remove Congo Red from contaminated water (Dhamin and Majeed, 2022; Gopinathan et al., 2015).

Because of its affordability and substantial capacity, uncomplicated regeneration, and low energy requirements, the adsorption mechanism has been considered an attractive strategy for treating wastewater. The type and characteristics of the used reactive material, though, have a significant

impact on effective the adsorption mechanism. Compared to other techniques, adsorption has several advantages, including applicability to toxic compounds, efficient removal of organic contaminants, flexible design and operation, and a smaller area requirement than biological systems (Majeed, 2017). Activated carbon may be expensive to use and regenerate, though, which makes it impractical in some cases. As a result, there has been an extensive study of cheaper and easily available sorbents in recent years. To treat wastewater contaminated with dyes, a variety of materials have been used, including clay minerals, peat, industrial byproducts, cement kiln dust, waste foundry sand, metal hydroxide sludge, and agricultural waste. Moreover, the ability of non-conventional sorbents to remove Congo Red from aqueous solutions has been researched which include clay, sludge biomass, and rice husk (Salleh et al., 2011; Wang et al., 2019).

Researchers across the globe are currently exploring the development of adsorbents using urban sewage sludge to create efficient and economically viable treatment methods for ecosystems, particularly through adsorption processes (Alshammari et al., 2020; Aoulad El Hadj Ali et al., 2021; Al Juboury et al., 2020). The impetus for this research direction stems from increasing apprehension over the release of organic compound-laden wastewater into the environment. The situation has been compounded by the swift expansion of specific human and industrial pursuits, such as the textile sector. Waterworks and sewage sludge are two products that are obtained from wastewater treatment, but they differ in composition and properties. Waterworks are facilities that utilize several purification steps, including coagulation, sedimentation, filtration, and disinfection, to make raw water safe for human consumption. The final product is clean and safe water that can be used for various domestic purposes. Conversely, sewage sludge is a solid residue that remains after the treatment of sewage, containing both organic and inorganic materials, including bacteria, viruses, pathogens, nutrients, and heavy metals. While sewage sludge may not be safe for direct use due to the associated bacteria, viruses, and pathogens generated during the biological treatment process, it can be further treated and processed to remove contaminants, resulting in a valuable resource for agricultural purposes (Faisal et al., 2020a). Moreover, upcycling waterworks sludge can aid in accomplishing

sustainability targets. As raw water resources for waterworks are often sourced from clay-rich rivers or natural water systems, the sludge derived from this process shares comparable traits to clay. As a result, it mainly consists of inorganic components and chemically introduced aluminum or iron during coagulation. Waterworks sludge possesses numerous advantageous characteristics, including an adverse surface charge, substantial potential for ion exchange, and porosity that facilitates the removal of a diverse range of contaminants, such as dyes.

Water treatment plants in Europe generate millions of tons of waterworks sludge annually, which is classified as “non-hazardous” under EU regulations, allowing for its disposal in sanitary landfills without additional costs. However, direct dumping of sludge into rivers can lead to increased water turbidity and higher filtration costs for potable water production. Water providers are currently actively searching for cost-effective solutions to tackle these concerns. The use of specific materials for eliminating colors from aqueous solutions has limits, especially in terms of cost and sorption capability, according to previous research. As a result, a new field of scientific study has developed focused on the manufacture of more effective materials with increased longevity and cheaper prices. Layered double hydroxides (LDHs) have attracted a lot of interest because of their potential uses in a variety of industries, including environmental remediation. LDHs are relatively inexpensive and straightforward to synthesize, making them viable for laboratory and industrial-scale production. These compounds offer an attractive option as reactive media for a wide range of chemical contaminants (Ahmed et al., 2020). The main goal of this investigation was to manufacture a sorbent from waterworks sludge that had been coated with (Mg/Al)-layered double hydroxide in order to eliminate CR from contaminated water. To determine the impacts of various manufacturing and operational factors, such as contact time, temperature, pH, and adsorbent dosage, the efficiency of this sorbent was investigated in batch tests.

## EXPERIMENTAL WORK

### Materials and methods

The study focused on Congo red (CR) dyes purchased from HIMEDIA (Indian). At room

temperature, a stock solution containing 1000 mg/L of dye was produced by mixing 1 gram of CR with 1 liter of distilled water. Various dye concentrations were achieved in water samples through dilution, and pH adjustments were carried out using 1 M NaOH or HCl with a German Bench Model Type WTW instrument. To evaluate the coated waterworks sludge, the dyes' effectiveness in treating a wastewater stream contaminated with these substances was assessed. The sludge utilized in this investigation was obtained from the AL-Weihdaa wastewater treatment facility situated in Baghdad, Iraq. The sludge obtained from the sedimentation tank was then severely air-dried, crushed, and milled into a powder.

### Manufacturing of reactive materials

The objective was to enhance the binding capacity and introduce new functional groups to the Waterworks sludge by depositing (Mg/Al)-LDH nanoparticles onto its surface. The resulting composite would serve as a low-cost immobilization matrix for use as a sorbent. To manufacture the nanoparticles, 25 ml of  $Mg(NO_3)_2 \cdot 6H_2O$  and 0.05 M  $Al(NO_3)_3 \cdot 9H_2O$  were employed, and  $NH_4OH$  (1 M) was gradually added to adjust the pH to 9, 10, and 11 with varying concentrations of Waterworks sludge. The pH was 6.5–7 after cleaning with distilled water and agitating the resulting adsorbent (Edmund Buhler SM25, German) for three hours to remove any unattached oxide.

### Batch tests

Conducting batch studies can determine the optimal operating conditions for achieving the highest possible removal of CR from contaminated water using reactive materials. These testing is used to determine variables such concentration of CR ( $C_o$ ), contact time, quantity of the sorbent, initial pH, and temperature. For the experiments, 250 mL flasks are required. In each flask contaminated water (50 mL) is mixed with a range of sorbent masses at different initial concentrations (10–300 mg/L) at 200 rpm for a contact time of 3 hours. Filter paper No. 1 by Wattmann is used to filter the adsorbent from the contaminated water, and a UV spectrophotometer is used to measure the concentration ( $C_e$ ) of the CR (PG Instruments, Model UV T80, England). The dye amount of absorbed by the sorbent ( $q_e$  mg/g) can be calculated using the principle of mass balance (Eq. 1). With

a dosage of sorbent between 0.1–2 g/50 mL and a contact time of three hours, the sorption tests may be carried out at a pH range of 3–12. (Faisal et al., 2020b; Lucas et al., 2004; Najji et al., 2020; Qiu et al., 2009; Weber and Morris, 1962).

$$q_e = (C_o - C_e) \frac{V}{m} \quad (1)$$

The sorption data is displayed between  $q_e$  and  $C_e$ , however by using Eq. 2, one may determine the adsorption efficiency (R).

$$R = \frac{(C_o - C_e)}{C_o} \times 100 \quad (2)$$

### Kinetics, equilibrium data and modeling studies

#### *Isotherms model*

To describe the relationship between the amount of adsorbate molecules adsorbed on the surface of the adsorbent and the concentration of CR in the liquid state at a specific temperature, sorption procedures often use an isotherm. The isotherm is a graphical representation of the correlation between the amount of adsorbate per unit mass of adsorbent and the equilibrium concentration of adsorbate in the solution ( $q_e$ ). To enable a standardized comparison of two different materials, the quantity is often normalized by the quantity of adsorbent utilized (Weber and Morris, 1962). Although increasing the adsorbate concentration may increase the quantity of adsorbed material per mass of adsorbent, the relationship between the two is not always linear (Perry and Chilton, 1984). The ideal isotherm should be convex upward and allow for a relatively high solid content at a low liquid concentration. A variety of adsorption isotherm models have been developed to analyze sorption data in this investigation.

1) The Friedrich model – in 1894, Freundlich and Kuster introduced the first nonlinear isotherm, known as the Friedrich model, The following can be used to illustrate the statement made the previous section: (Qiu et al., 2009).

$$q_e = K_f C_e^{1/n} \quad (3)$$

where: the Freundlich constants,  $K_f$  and  $n$ , respectively represent the amount and intensity of adsorption.

2) Langmuir's model – the Langmuir isotherm, invented by Langmuir in 1916, is commonly

employed to characterize the transfer of chemicals between liquid and solid phases. In this model, the solute flows within the surface with no change in the adsorption energy, and the sorption process occurs when a monolayer of saturated solute molecules occupies the sorbent surface. Equation 4 depicts the formal expression of the Langmuir isotherm (Qiu et al., 2009).

$$q_e = \frac{q_m b C_e}{1 + b C_e} \quad (4)$$

#### Kinetics model

To accurately predict the rate at which a solute is eliminated from water, it is crucial to understand the sorption mechanism and the pace at which sorbate molecules are assimilated at the interface (solid/solution). Kinetics plays the vital role for determining the solute adsorption rate and the optimal duration for effective sorption treatment. Both of the kinetic models presented below are necessary to fully understand the sorption process and identify the predominant stage (Yagub et al., 2012):

1) Pseudo-first-order kinetics – Lagergren introduced a kinetic rate for the first-order adsorption process in either the liquid or solid phase, which is dependent on the solid's adsorption capacity. The quantitative characteristics of this concept are analyzed in the following text (McKay et al., 1999).

$$q_t = q_e (1 - e^{-k_1 t}) \quad (5)$$

The maximum adsorption capacity ( $q_e$ ) of the sorbent is measured in mg/g, while the amount of sorbate present at any given time ( $q_t$ ) is also measured in mg/g. Additionally, the pseudo-first-order rate constant ( $k_1$ ) is expressed in  $\text{min}^{-1}$  and remains constant throughout the adsorption process.

2) A pseudo-second-order kinetics model – was developed using the following assumptions (Saha et al., 2018):

- a) sorbate particles form a monolayer on the surface of the sorbent;
- b) all sorbent particles possess the same adsorption energy irrespective of the degree of surface coverage; and
- c) adsorption occurs at distinct sites where the sorbate being adsorbed does not interact, and the adsorption rate is insignificant compared to other processes. The equation below represents this model:

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

where:  $q_t$  – represents for the quantity of contaminant adsorbed at time  $t$  (mg/g);

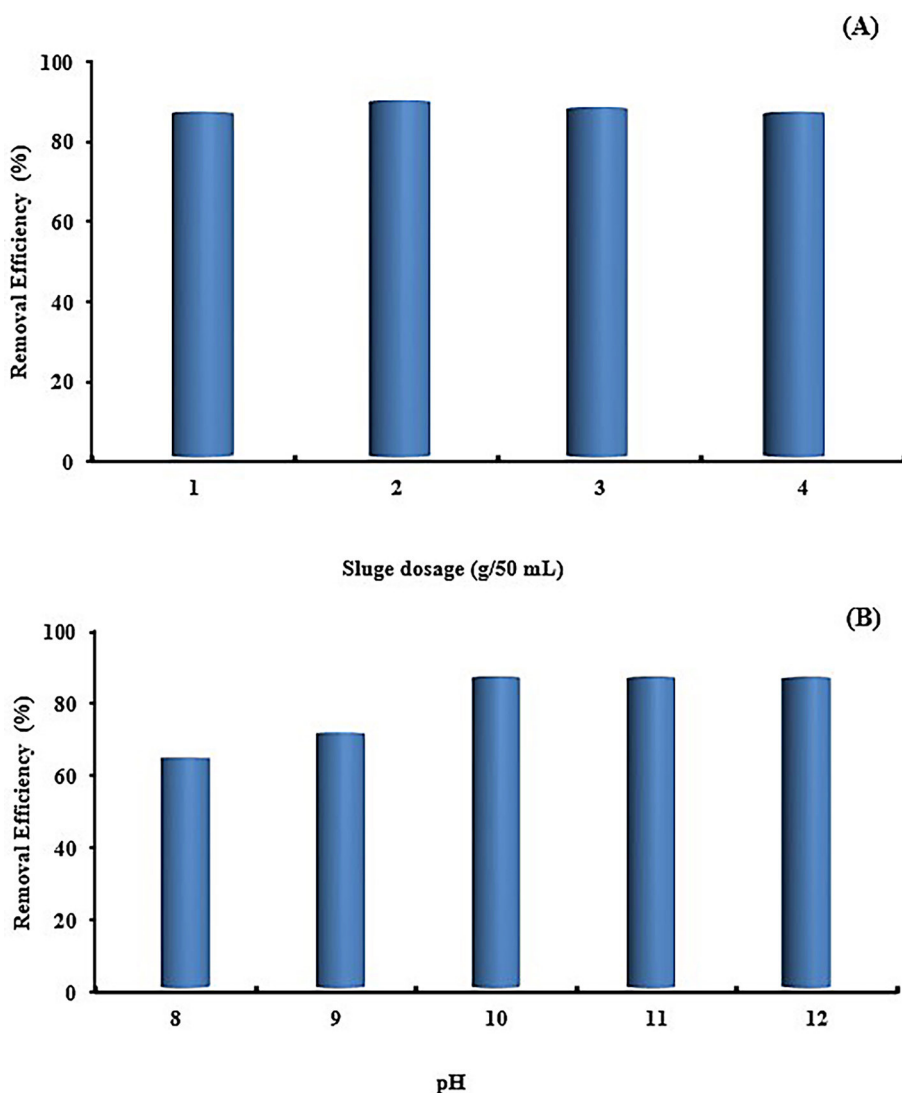
$k_2$  – the second-order rate constant (g/mg min).

## RESULTS AND DISCUSSION

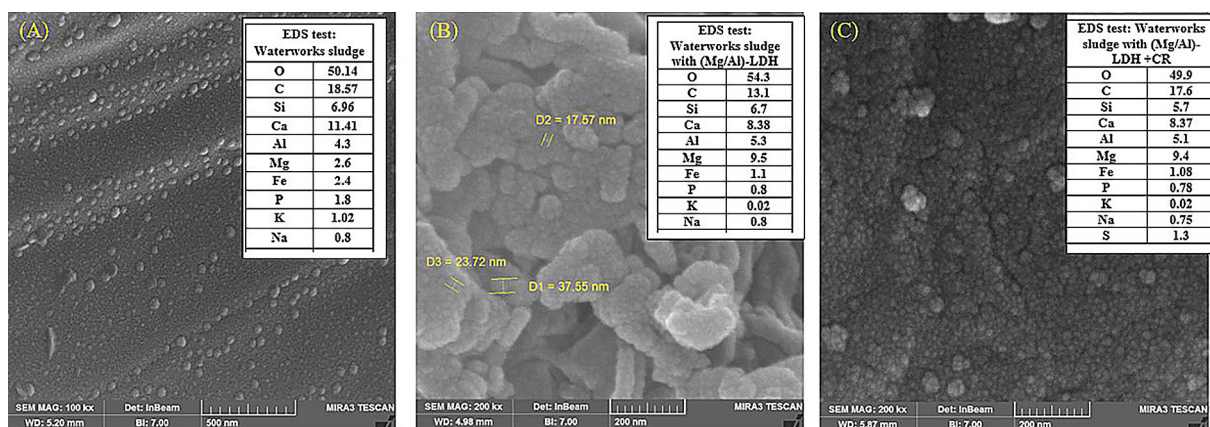
### Manufacturing coated waterworks sludge with MgAl-LDH

The effects of two factors, solution pH and sludge dosage, must be examined in order to determine the removal of the sorbent and the best values for the aforementioned parameters in order to produce waterworks sludge covered with (Mg/Al)-LDH. The experimental conditions involved the use of 0.5 g of adsorbent in 50 mL of solution, with a concentration of 50 mg/L of CR, a pH of 3, and a contact time of 3 hours under agitation at 200 rpm. Five pH values (8, 9, 10, 11, and 12) were tested, and the sorption capabilities were found to be similar. Figure 1 shows the relationship between pH and the sorbent's removal efficiency for CR dye adsorption. The results indicate that surface modification through (Mg/Al)-LDH has a significant effect on removal compared to virgin waterworks sludge, which stabilizes at 59% for CR. Based on the graph, it can be observed that a pH value of 10 resulted in a removal effectiveness of approximately 88.25%. A significant reduction in removal efficiency may occur if the pH deviates from 10, possibly due to incomplete nanoparticle adhesion to the surfaces of the waterworks sludge or an increase in nanoparticle diameter (Ma et al., 2019). Therefore, the optimal coating procedure can be achieved at pH 10. The impact of waterworks sludge dosage (adjusted from 1 to 4 g/50 mL) on the sorbent generation at the previously mentioned pH is examined, and the outcomes are presented in Figure 2. Based on the graph, adding 2 g to 50 mL of the solution can yield a sorbent that exhibits a removal efficiency of 91.19%, indicating its potential to bind a significant portion of CR dyes. However, excessive addition of waterworks sludge would result in a clear decline in the removal efficiency, as the additional nanoparticles cannot adequately cover the increase in surface area when compared





**Figure 1.** pH (A) and waterworks sludge dosages (B) effect on the synthetic process of waterworks sludge that has been coated with (Mg/Al)-LDH nanoparticles for treating water contaminated with CR dyes (dosage = 0.5 mg/50 mL, C = 50 mg/L, T = 25 °C, time = 3 h, pH = 3).



**Figure 2.** Scanning electron microscopic (SEM) images and EDS elemental composition analysis for waterworks sludge (A), waterworks sludge coated with (Mg/Al)-LDH (B), Congo red, and sorbent after adsorption (C)

to optimal dosages. Moreover, for lower doses of waterworks sludge, it is likely that many nanoparticles will not get an opportunity to adhere to the surfaces of the sludge and will be washed away after the preparation process.

### SEM-EDS analysis

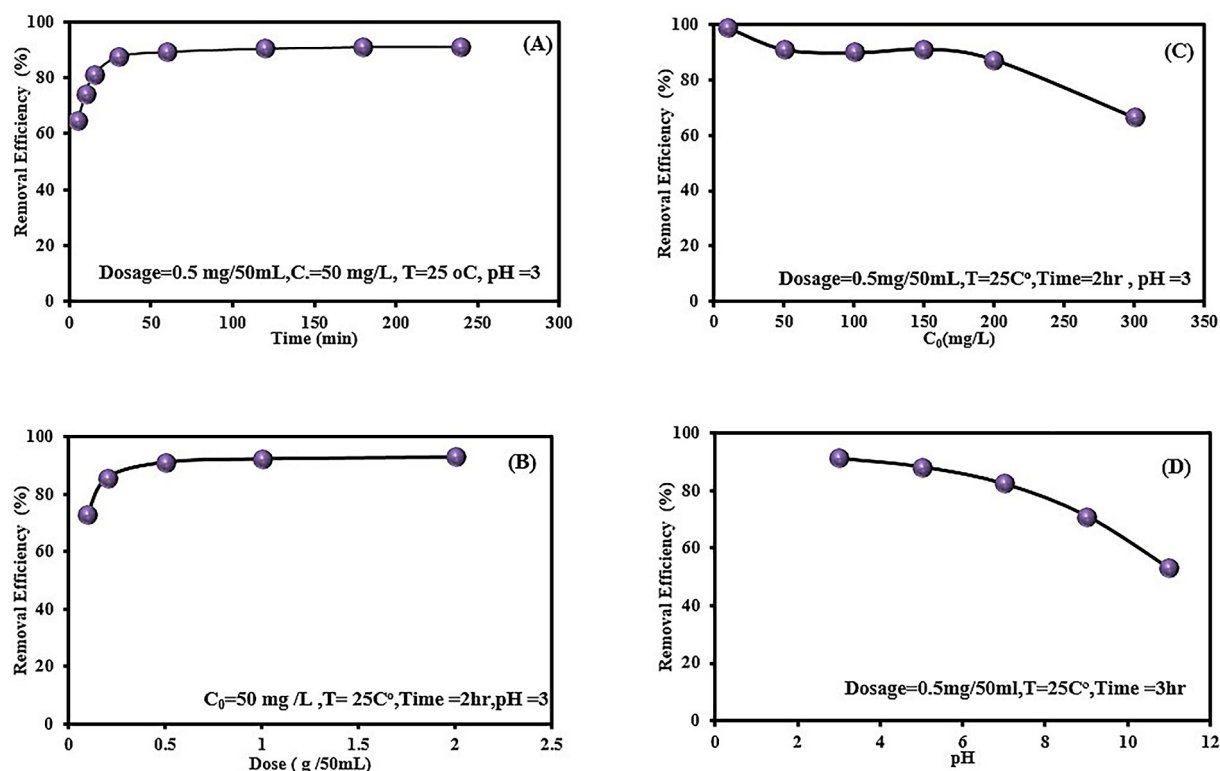
Energy-dispersive X-ray spectroscopy (EDS) was utilised to construct elemental compositional graphs for waterworks sludge coated with (Mg/Al)-LDH, Congo red, and sorbent, as depicted in Figure 3. As depicted in the figure, the composition analysis reveals a notable presence of O, C, Si, Ca, Al, Mg, Fe, P, K, Na, and S in the waterworks sludge when combined with EDS. The table indicates that the application of (Mg/Al)-LDH coating on the waterworks sludge significantly increased the proportion of Mg and Al, indicating successful implantation of the (Mg/Al)-LDH onto the sludge. The findings indicate a significant rise in the carbon percentage of the adsorbent from 13.1% (prior to sorption) to 17.6%, implying the presence of adsorption. Remarkably, the utilization of the adsorbent post-sorption enabled the efficient elimination of Congo red (CR) from solutions. Figure 3

displays the characteristics of the waterworks sludge before and after the coating process, as observed through SEM. The images unmistakably illustrate the emergence of crystals on the sorbent's surface, which is a clear indication of the formation of (Mg/Al)-LDH. However, the mechanism underlying the attachment of (Mg/Al)-LDH nanoparticles to the surface remains uncertain. Notably, there are noticeable differences in the morphology of the sorbent before and after the sorption of CR, possibly due to the binding of contaminant particles.

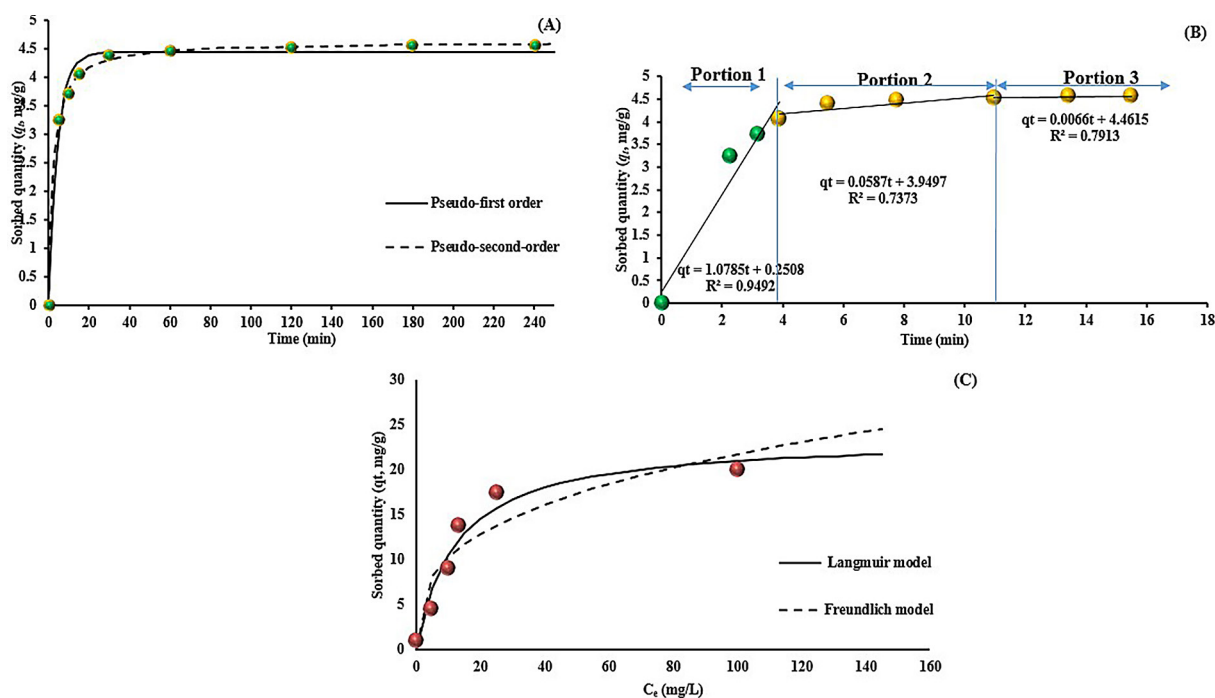
### Operating conditions effect

#### The effect of contact time

To ensure the attainment of equilibrium concentrations, a fixed contact time is necessary to be maintained during batch experiments. In an experiment carried out at 25 °C, 50 mL of a Congo red (CR) solution with an initial concentration ( $C_0$ ) of 50 mg/L and a pH of 3 were employed. The mixture was agitated at 200 rpm, and 0.5 g of waterworks sludge coated with (Mg/Al)-LDH nanoparticles was added. Figure 4 illustrates the relationship between the removal efficiency and contact time. The graph reveals that an increase in



**Figure 3.** The effect of contact time (A), dosages (B), initial concentration (C), and pH (D) on the efficiency of CR removal in batch experiments



**Figure 4.** Experimental data was utilized to analyze the sorption kinetics (A), intra-particle diffusion model (B), and isotherm models (C) of CR onto coated waterworks sludge

contact time leads to a significant reduction in the amount of CR. The rate of adsorption first rises and then gradually falls over time, indicating a gradual loss of vision in adsorption capacity of the adsorbent surface's. Around 90.6% of CR was gone in two hours, and the residual concentrations of CR remain at a constant level, with only minor changes for up to two hours. The high adsorption rate is attributed to the sorbent surface's capacity for reactive sites and the declining rate tendency over time due to CR particles occupying the active sites. The sorption findings are consistent with previous studies, such as Othmani et al., (2013) work on the removal of CR on altered clay with nan-montmorillonite and Faisal et al., (2021) research on the trend of interactions comparing Iraqi bentonite with CR. Therefore, it be appropriate to use a contact time of two hours for sorption tests, as it appears to be adequate to achieve the equilibrium state.

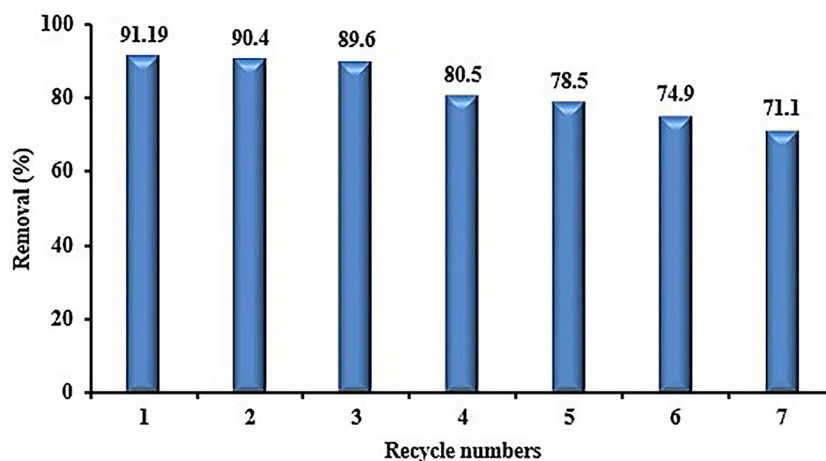
#### The effect of CR concentration

Additional experiments were carried out to explore how changes in the initial concentrations ( $C_0$ ) of CR affect the effectiveness of sorption. The employed  $C_0$  concentrations varying between 10 and 300 mg/L, an initial pH of 3, adsorbent incorporation, and stirring for two hours at 200 rpm. The results are presented in Figure 5, which

exhibits the equilibrium elimination rates of CR. The findings indicate that the effectiveness of removal increases with increasing  $C_0$  concentrations up to 300 mg/L, but declines for lower  $C_0$  values. The observed changes can be attributed to the saturation of the active sites of the sorbent, which bind to the CR contaminants. Furthermore, the results indicate that a greater quantity of CR contaminants are adsorbed per unit mass of the sorbent at equilibrium when the initial concentration ( $C_0$ ) is lower, with the amount of dye adsorption increasing as the concentration rises. This phenomenon strengthens the affinity between the contaminants and the binding sites, promoting the interaction between the adsorbent and CR. As the initial concentration increases, the removal rate decreases. The production of (Mg/Al)-LDH nanoparticles necessitated the presence of  $H^+$  and  $OH^-$  in the aqueous solution for protonation and deprotonation to occur. The exterior of (Mg/Al)-LDH nanoparticles contains two different reactive sites, namely Mg-OH and Al-OH. Therefore, the polar or ionic groups of contaminants interact with these functional groups in various ways (Othmani et al., 2013).

#### Effect of sorbent dosage on the sorption process

In order to explore the effect of adsorbent dosage on CR adsorption, coated waterworks sludge



**Figure 5.** Congo red dye removal efficiency from aqueous solution using waterworks sludge coated with (Mg/Al)-LDH in recycle tests at room temperature (dosage = 0.5 mg/50 mL, T = 25 °C, time = 3 h)

was employed at various dosages ranging from 0.1 to 2 g. Each dose was mixed with a 50 mL solution of CR at 25 °C, 200 rpm, and a contact time of 2 hours, with an initial pH of 3. As shown in Figure 6, the removal efficiency is increased by increasing the adsorbent dosage within the selected range, as more active sites become available for adsorption, as expected.

#### Effect of pH solution

The pH is considered the most significant variable as it controls the degree of ionization of CR, the charge on the sorbent surface, and the dissociation of functional groups on the sorbent (Pengthamkeerati et al., 2010; Wawrzkievicz and Hubicki, 2009). As shown in Figure 7, the efficiency of CR removal decreases from 52.95% to 91.19% with an increase in pH from 3 to 11. The lowest removal percentage was observed at pH 11. However, since neutral conditions represent large-scale circumstances and the removal at pH 7 is unaffected by changes in acidity, pH 7 is recommended as the ideal pH value for the solution. At pH 7, the anionic dye can form strong electrostatic interactions with the positive surface of the sorbent, thereby enhancing dye sorption. Under neutral conditions, the hydration and ionization of the dye can decrease, which can improve the removal process through hydrogen bonding and  $\pi$ - $\pi$  stacking effects. Conversely, as the pH decreases, the positive charges on the surface of the sorbent increase, leading to a negatively charged exterior that repels the dye and makes its removal more challenging (Puranik et al., 1999). The reduction in sorption at higher pH values can be attributed to the competition

between hydroxyl ions and the dye for the available binding sites. The decrease in efficiency at higher pH levels may be due to the generation of OH<sup>-</sup> ions, leading to a reduction in hydrogen bonding. Furthermore, alterations in the distance between functional groups on the sorbent's surface in response to pH variations can also influence the sorption process's equilibrium.

#### Sorption kinetics

The sorption kinetics, which influences the equilibrium time, determines the rate of CR adsorption onto the sorbent particles. Modeling the sorption process can help identify the adsorption mechanism that governs the movement of contaminants from an aqueous solution to the sorbent material. The sorption process simulation depends on the sorbent's surface-level reactions. Among the preferred kinetic models for sorption are the pseudo-first-order and pseudo-second-order kinetic expressions. Predicting the rate of sorption of dissolved pollutants from the liquid phase is crucial for developing effective treatment techniques (Wawrzkievicz and Hubicki, 2009). The experimental data of CR sorption onto the adsorbent were analyzed using kinetic models, as presented in Figure 8. The solid line corresponds to the pseudo-first-order model, while the dashed line represents the pseudo-second-order model. Table 1 shows the parameters for these models, which were obtained through nonlinear regression using Excel 2016's "solver" function. Based on the R<sup>2</sup> and best qe fit values to the test data, it can be concluded that the pseudo-second-order model is the most



appropriate method for describing the sorption process of the investigated pollutants. This indicates that the primary mechanism involved is chemisorption, and that the exchange of electrons between the active sites of the sorbent and the sorbate may reflect valence forces that determine the rate of sorption (Komy et al., 2014). In order to gain a deeper comprehension of sorption mechanisms, it is recommended to combine kinetic models with the Weber and Morris diffusion model (1962). This type of Adsorption volume  $t^{0.5}$  value is tied as follows, instead of using  $t$ , to describe the empirical equation.

$$q_t = k_{int} t^{0.5} + C \quad (7)$$

To describe the mechanisms involved in sorption processes, combining kinetic models with Weber and Morris' diffusion model (1962) is essential. Figure 8 presents the straight-line relationship for the examined pollutants with relatively good  $R^2$  values between  $q_t$  values and  $t^{0.5}$  straight-line plots. This indicates that diffusion of intra-particles is taking place in the process of adsorption even when particles are not going through the source, even though the rate-regulating step may not be displayed. Based on the Figure, it can be inferred that there are multiple simultaneous processes involved in CR adsorption that are under regulation, as indicated by the observed multi-linearity in the intra-particle diffusion plots.

### Sorption isotherms models

The use of the sorption isotherm enables the characterization of the distribution of pollutant particles between the liquid and solid phases at equilibrium, as well as the determination of the adsorbent's maximum adsorption capacity and affinity. To interpret the experimental results, the Freundlich and Langmuir models of sorption isotherms were employed. Under optimal conditions, a range of carefully selected contaminants with concentrations ranging from 10 to 300 mg/L were subjected to adsorption tests. A variety of sorbents, including coated waterworks sludge, were prepared and used in these tests. The sorption measurements were analyzed using nonlinear regression by utilizing the solver function in Microsoft Excel 2016 and the Freundlich and Langmuir isotherm models. Table 1 presents the sum squared error (SSE) and coefficient of determination ( $R^2$ ) values

for these model systems. It is abundantly evident that the Langmuir model outperforms the Freundlich model in its ability to explain experimental data, with CR Langmuir constants for coated waterworks sludge being 0.08 l/mg. Figure 9 shows how closely the sorption isotherm models and experimental findings agree. The maximal capacity for adsorption of the coated waterworks sludge for CR in a solution is shown in Table 2, indicating a significantly higher adsorption capacity (23.57 mg/g) The stronger interaction between CR particles and the adsorbent surface is responsible for this.

### Recyclability

To evaluate the practical utility of an adsorbent, its capacity to maintain performance and regenerate during the adsorption process is crucial. To regenerate the used adsorbent, a 0.1M NaOH solution was utilized to eliminate the adsorbed Congo red molecules for three hours. The three-time recycling tests presented in Figure 5 reveal that the sorbent efficiently removes Congo red dye with an efficacy exceeding 89%. The results suggest that the (Mg/Al)-LDH nanoparticle-coated waterworks sludge is a promising adsorbent for treating water contaminated with anionic dyes.

### CONCLUSIONS

The research demonstrated that coating (Mg/Al)-LDH nanoparticles onto waterworks sludge can effectively extract CR from contaminated aqueous solutions. The batch trials yielded dependable results, indicating the potential to use solid waste in the remediation process in line with environmental and sustainable development goals. The Langmuir isotherm model, with an  $R^2$  value of 0.956 and a maximum adsorption capacity of 23.576 mg/g, outperformed the Freundlich model in predicting sorption values.

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