

# Comparison studies of Double Layered Hydroxides modified by sodium dodecyl sulphate and Graphene Oxide composites for the treatment of Coomassie blue (G-250) in wastewater

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**Abstract:** In this study, Sodium Dodecyl Sulphate modified double layered hydroxides (SDS-LDH) and Graphene Oxide Double Layered Hydroxides (GO-LDH) were synthesized via reconstruction and sonochemical methods, respectively. Batch experiments were performed to study the adsorption behaviour and mechanism of the synthesized adsorbents to eliminate Coomassie brilliant blue G-250 dye (CBB) from an aqueous solution. GO-LDH was more efficient in CBB adsorption through ion exchange, resulting in  $q_e = 1430$  mg/g using 5 mg of GO-LDH at 5 min owing to its high specific average surface area and inter-lamellar distance. However, SDS-LDH depended on the hydrophobic interaction with CBB leading to  $q_e = 62$  mg/g using 85 mg of SDS-LDH at 60 min. SDS-LDH and GO-LDH obeyed the PSO model with a GO-LDH adsorption initial rate 7 times higher than that of SDS-LDH as discussed by the intra-particle diffusion model via three steps.

**Keywords:** SDS-LDH; GO-LDH; CBB (G-50); Harkins-Jura; Langmuir

## 1. Introduction

The world has recently been afflicted by serious water pollution. Textile, printing, and painting industries resulted in dye pollution (Abbas et al., 2016). Dyes are not only harmful to living things because of their intense colors, toxicity, mutagenicity, and carcinogenicity but also difficult to remove due to their complicated aromatic structures; water-solubility, anti-oxidative properties; and low degradability (Tang et al., 2023), (Sun et al., 2020). The evaporated chemicals caused by dyes in wastewater might affect climate change, resulting in respiratory infections, human organs damage, killing the aquatic life, increasing drought in some areas, and decreasing rain fall in other areas (Jawad et al., 2021). Various techniques were introduced for the wastewater treatment, such as: catalytic degradation (Tabana et al., 2023), advanced oxidation (Gupta et al., 2023), photocatalysis (Gohr et al., 2019), electrocatalysis (Liu et al., 2015), membrane separation (Zhao et al., 2023), and adsorption (Gohr et al., 2022). The adsorption process was the most successful method due to its simplicity, lack of environmental pollution, and low cost (Yilmaz, 2022). The anionic organic dye Coomassie brilliant blue G-250 (CBB) (triphenylmethane) is well-known for its role in protein determination and the wool industry (Lü et al., 2007), (Rayaroth et al., 2015), however it is harmful to humans, causing mucus membrane irritation, respiratory tract toxicity (Tian et al., 2023), and eye problems (Sujitha & Ravindhranath, 2016). CBB could be removed by the adsorption process utilizing numerous adsorbents, including active carbon derived from barks of Ficus racemosa plant (Sujitha & Ravindhranath, 2016), amino-functionalized silica smart nano particles (Sabeela et al., 2019), starch/poly (alginic acid-cl-acrylamide) nanohydrogel (Sharma et al., 2017), activated carbon prepared from Nigella sativa L. waste (Abdel-Ghani et al., 2017), snail shell powder (Ibrahim et al., 2019), wheat bran (Ata et al., 2012), apricot stone activated carbon (Abbas et al., 2016),  $\alpha$ -chitin nanoparticles (Dhananasekaran et al., 2016), sand (Rauf et al., 2008), Fe-Pillared bentonite (PILC-Fe), Benta-Na (Kadeche et al., 2020), and CuO/C nanocomposites (Bhavyasree & Xavier, 2021). Double Layered Hydroxides (LDH-s) are considered as anionic clays (2d-materials),  $[M_{1-x}^{2+}M_x^{3+}(\text{OH}_2)]^{x+}(\text{A}^n)_{x/n} \cdot n\text{H}_2\text{O}$ , that made up of positive cation layers ( $M_{1-x}^{2+}M_x^{3+}$ ) bonded to hydroxyl groups, intercalated with negatively charged inorganic anions ( $\text{A}^n$ )<sub>x/n</sub> and water molecules ( $\text{H}_2\text{O}$ ), neutralizing its lamellae. LDH exhibited ion exchange, complexation, surface adsorption, good regeneration efficiency, high specific surface area, and no health hazard (Rashed et al., 2022), (Daud et al., 2019). Furthermore, LDH layers could be intercalated with organic molecules rather

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than inorganic molecules, resulting in hydrophobic character, enhancing organic pollutant adsorption (Starukh et al., 2016). LDH-s can be synthesized using a variety of methods (Rashed et al., 2022). According to our previous work (Rashed et al., 2022), ZnAl-CO<sub>3</sub> LDH was used to remove Methyl Orange (MO) and CBB from an aqueous solution, but the adsorption capacity did not exceed 7.5 mg/g, which might be due to the difference in size and charge of the intercalated anion (CO<sub>3</sub><sup>2-</sup>) corresponding to LDH interlamellar distance and that of CBB during ion exchange adsorption (Elmoubarki et al., 2021). Moreover, graphene oxide (GO) was chosen for the adsorption process due to its highly concentrated active sites (C-OH, C=O, COOH, C-O-C) forming hydrogen bonds from the inner outer layers and high specific area that led to adsorbing pollutants as heavy metals, pesticides, endocrine disrupting compounds, phenols, chlorophenols, and dyestuff from wastewater (Abu-Nada et al., 2021). It was reported that GO led to increasing the adsorption capacity due to the wide specific area of hydrogen bond interactions between GO and CBB, but only 14.5 mg/g was achieved due to the repulsion between the negative functional groups of oxygen of the GO surface and the CBB anions (Abu-Nada et al., 2021). Herein, the LDH was synthesized using a modified reconstruction method by incorporating the organic surfactant sodium dodecyl sulphate (SDS) after calcining LDH at 500°C, yielding (SDS-LDH) of hydrophobic nature (Starukh et al., 2016). Additionally, the LDH lamellae was sonochemically grown into GO layers forming (GO-LDH). The two modified synthesized adsorbents were expected to enhance the CBB dye removal. The synthesized adsorbents were characterized and their adsorption parameters, kinetic models, isotherms, thermodynamics were assessed. Besides, recycling using the memory effect process to study the mechanism of both adsorbents towards CBB adsorption for eventual use was performed (Starukh & Levytska, 2019), (Mishra, et al. 2018).

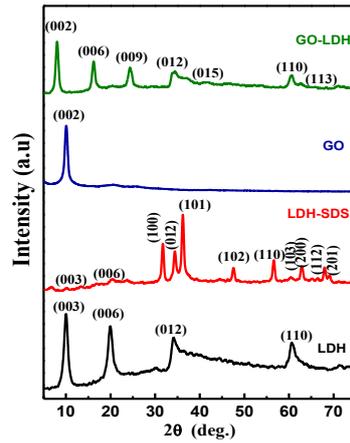
## 2. Methodological Framework

The used resources, preparation of the adsorbents, characterization techniques and are mention in the supplementary data in detail.

## 3. Results and Discussion

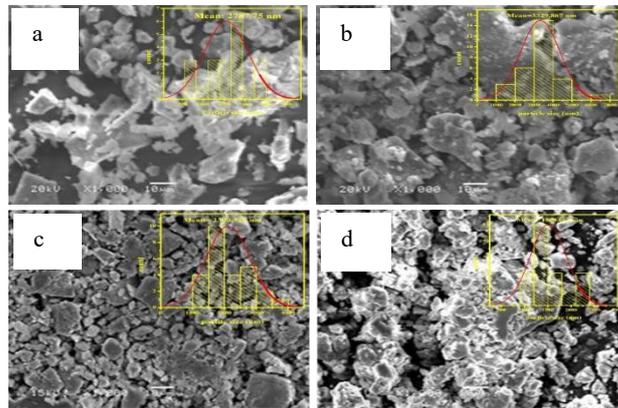
### 3.1. Physicochemical characterization of SDS-LDH and GO-LDH composites

Fig. 2 displayed the XRD patterns of LDH, SDS-LDH, GO and GO-LDH. The XRD pattern of LDH exhibited the basal peaks (003) and (006) at  $2\theta$  of 9.9455° and 19.936° and the non-basal peaks (012) and (110) at  $2\theta$  of 34.15° and 60.61°, respectively, referring to the good crystallinity of LDH layers. It was found that after the formation of SDS-LDH composite (003) and (006) basal peaks were shifted to higher ( $2\theta$ ) of 10.04° and 20.4° and their peaks intensities were significantly decreased due to the absence of CO<sub>3</sub><sup>2-</sup> interlayered anion (Abd-Elhamid et al., 2019), (Mahjoubi, et al. 2019). Additionally, the intensity of (012) and (110) non-basal peaks increased, indicating the enhanced crystallinity of LDH by the organic anion SDS (Ramimoghadam, et al., 2012). Besides, (100), (101), (102), (103), (200), (112), and (201) peaks appeared at ( $2\theta$ ) of 31.5°, 36.22°, 47.78°, 60.1°, 62.71°, 67.97°, and 70.11°, respectively owing to the perfect orientation of SDS with ZnO (Mahjoubi, et al, 2019). Therefore, SDS-LDH adsorbed CBB dye via hydrophobic interactions with SDS (Mahjoubi, et al, 2019). Moreover, in the XRD pattern of GO-LDH, the GO characteristic peak at  $2\theta$  of 10.0115° disappeared due to GO sheets dispersion between LDH layers as a result of the electrostatic attraction between the LDH metals and the oxygen functional groups of GO sheets (Rashed et al., 2022). The basal peaks (003) and (006) were also shifted to  $2\theta$  of 8.0061° and 16.094°, respectively, along with strengthening and sharpening their intensities, revealing CBB adsorption easily via anion-exchange due to high interlayered distance. In addition, new non-basal peaks (009), (015), and (113) were observed at  $2\theta$  of 25.44°, 35.54°, and 61.72°, respectively, referring to the excellent crystallinity due to the well-grown ZnO on GO sheets (Rashed et al., 2022).

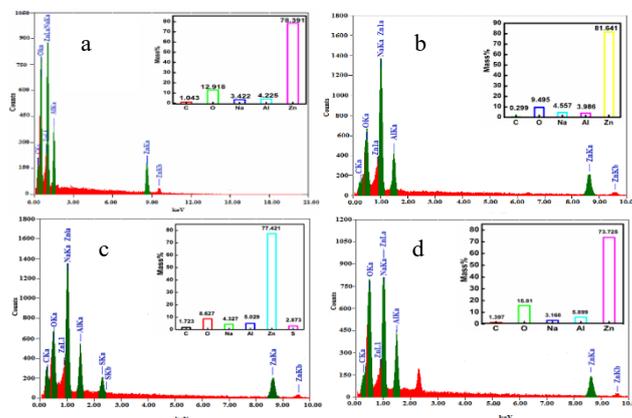


**Fig. 2: XRD patterns of LDH, SDS-LDH, GO and GO-LDH.**

The morphological investigation and EDS spectra of LDH, C-LDH, SDS-LDH and GO-LDH were displayed in Fig. 3 and Fig. 4, respectively. Their corresponding specific area distribution curves were shown in Fig. S1, supplementary data. The SEM images depicted the layered arrangement of LDH with an average particle size of 2787.75 nm (Fig. 3a) and an average specific surface area of  $21.2 \times 10^6 \text{ nm}^2/\text{g}$  (Fig. S1a, supplementary data). After calcination, C-LDH morphology exhibited flakes with rough surface with an average particle size of 3329.867 nm (Fig. 3b) and a lower average specific surface area of  $16.7 \times 10^6 \text{ nm}^2/\text{g}$  (Fig. S1b) due to the removal of  $\text{H}_2\text{O}$  and  $\text{CO}_3^{2-}$  upon calcination (Starukh et al., 2016). SEM images of SDS-LDH possessed more dispersed layers of an average particle size of 2300.526 nm (Fig. 3c), due to the strong affinity of LDH to organic surfactants other than  $\text{CO}_3^{2-}$  (Kong et al., 2018). Besides, SDS increased the average specific surface area of LDH to  $35.5 \times 10^6 \text{ nm}^2/\text{g}$ . Whereas, incorporation of GO to LDH perfectly improved the morphological arrangement of LDH layers (Fig. 3d) owing to the well-dispersed LDH metals onto GO sheets that led to a decrease in its average particle size to 1508.667 nm along with an increase in the average specific surface area to  $47 \times 10^6 \text{ nm}^2/\text{g}$  (Fig. 1Sd) (Rashed et al., 2022). The EDS results confirmed the presence of C, O, Na, Al and Zn elements in the synthesized composites, indicating the great accuracy of LDH synthesis. In case of C-LDH, both of carbon and oxygen contents decreased due to successful calcination process via  $\text{CO}_3^{2-}$  elimination (Fig. 4b). After the reconstruction of SDS-LDH, sulphur content appeared due to the sulphonyl group of SDS (Fig. 4c). Additionally, carbon content increased in SDS-LDH and GO-LDH as a result of the presence of the SDS alkyl chain and the carbon skeleton, respectively (Zhang et al., 2019).



**Fig. 3: The SEM image of a) LDH, b) C-LDH, c) SDS-LDH and d) GO-LDH.**

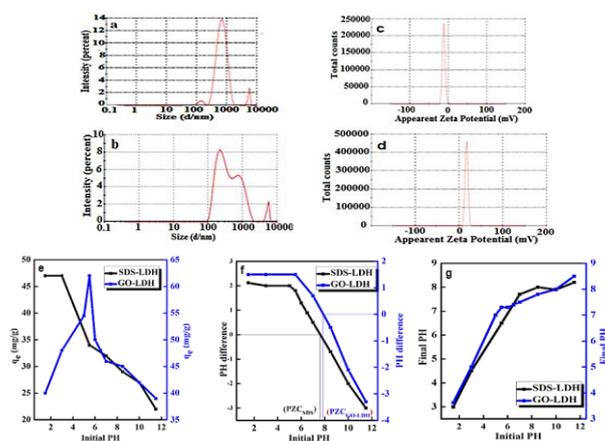


**Fig. 4: EDS spectrum of a) LDH, b) C-LDH, c) SDS-LDH and d) GO-LDH**

Infrared spectra of LDH, C-LDH, SDS-LDH, GO, GO-LDH, CBB/SDS-LDH and CBB/GO-LDH were shown in Fig. 5a. The FT-IR spectrum of LDH exhibited a wide band at  $3440\text{ cm}^{-1}$  due to the hydroxyl group vibration bond of M-O-H or H-O-H, a sharp band at  $1560\text{ cm}^{-1}$  of water molecule bending vibration, and a band at  $1385\text{ cm}^{-1}$  as a result of interlayered anion  $\text{CO}_3^{2-}$  stretching mode (Rashed et al., 2022). LDH calcination process led to a shifted narrow band at  $3432\text{ cm}^{-1}$  due to the elimination of OH hydroxyl groups from LDH forming C-LDH. The two sharp bands at  $1385$  and  $1560\text{ cm}^{-1}$  of LDH disappeared for C-LDH, proving successful calcination (Elmoubarki et al., 2021). The FT-IR spectrum of SDS-LDH exhibited sharp peaks at  $2919$  and  $2852\text{ cm}^{-1}$  due to C-H (alkyl chains) stretching and bending vibrations and peaks at  $1217$ ,  $1062$ , and  $990\text{ cm}^{-1}$  of  $\text{OSO}_3^-$  anions stretching and bending vibrations. Besides, the peaks at  $3480$  and  $1632\text{ cm}^{-1}$  of hydroxyl group stretching and water molecules bending vibrations were shifted and weakened in intensity as compared to LDH, which might be due to the decrease of the O-H bond of M-O-H or H-O-H of LDH as a result of the great affinity of LDH metals to bond with SDS anions (Starukh et al., 2016). The FT-IR spectrum of GO sheets displayed a characteristic peak at  $3453\text{ cm}^{-1}$  of O-H vibration of OH or COOH functional groups on GO surface, a band at  $1757\text{ cm}^{-1}$  of C-O vibration of COOH, and bands at  $1633$ ,  $1410$ , and  $1038\text{ cm}^{-1}$  of C-O-H, C-O-C, and C-O stretching modes of GO carbon skeleton, respectively (Rashed et al., 2022). After LDH growth on GO sheets (GO-LDH), the broad band due to the O-H vibration of hydroxyl groups and water molecules was shifted to  $3495\text{ cm}^{-1}$  as well as the bands due to the bending interlayered water molecules and  $\text{CO}_3^{2-}$  anions were shifted to  $1640$  and  $1385\text{ cm}^{-1}$ , respectively. The stretching modes of C-O-C and C-O-H were also shifted to  $1430$  and  $1100\text{ cm}^{-1}$ , confirming that LDH layers were perfectly constructed on the GO sheets. The shifted bands of metal oxides also proved that the metals were well-complexed with GO oxygen functional groups (Rashed et al., 2022). Illustratively, the bands observed at  $600 - 400\text{ cm}^{-1}$  were due to M-O-H and M-O-M vibrational modes of metal oxides of LDH, C-LDH, SDS-LDH, GO-LDH (Rashed et al., 2022), (Starukh & Levytska, 2019). After CBB adsorption by SDS-LDH, the 2 bands of the alkyl chains were shifted to  $2921$  and  $2854\text{ cm}^{-1}$ , indicating hydrophobic interactions of alkyl chains with CBB (Munir et al., 2020). On the other hand, CBB adsorption by GO-LDH exhibited a significant decrease in the peak intensity of  $\text{CO}_3^{2-}$  at  $1385\text{ cm}^{-1}$ , referring to that the interlayered space was embedded with CBB molecules. FT-IR spectra of CBB adsorption with either SDS-LDH or GO-LDH exhibited bands shift at  $3511$  and  $3462\text{ cm}^{-1}$  for the hydrogen bonds and at  $700 - 400\text{ cm}^{-1}$  for M-O-M and M-O-H metal oxide vibrations (Munir et al., 2020). The later shift indicated metal complexations with CBB dye for both SDS-LDH and GO-LDH (Rashed et al., 2022). The Raman spectra of the synthesized composites were displayed in Fig. 5b. For LDH, the band at  $1392\text{ cm}^{-1}$  due to stretching mode of interlayered  $\text{CO}_3^{2-}$  as well as the two split bands at  $1090$  and  $1060\text{ cm}^{-1}$  might be due to the formation of M-O symmetrical stretching bond in  $\text{M-CO}_3^{2-}$  (Rashed et al., 2022). The band at  $2860\text{ cm}^{-1}$  was due to O-H vibration between carbonate and water molecules (Rashed et al., 2022). Raman spectrum of C-LDH revealed that the carbonate band of LDH at  $1392\text{ cm}^{-1}$  was diminished due to the removal of  $\text{CO}_3^{2-}$ , which confirmed FTIR results. Additionally, the band at  $2860\text{ cm}^{-1}$  of LDH completely decreased in C-LDH due to the removal of O-H bonds between  $\text{CO}_3^{2-}$  and  $\text{H}_2\text{O}$  after the calcination process (Lin et al., 2014). SDS-LDH Raman spectrum exhibited only one sharp peak at  $2888\text{ cm}^{-1}$  due to SDS alkyl chain vibrational mode (Islam et al., 2013). The bands  $800 - 500\text{ cm}^{-1}$  were due to M-O-M and M-O-H metal oxides vibrations of C-LDH and SDS-LDH (Islam et al., 2013). Raman spectra of GO revealed 2 main bands: D-band at  $1362\text{ cm}^{-1}$  due to disordered graphitic carbon defects and G-band at  $1584\text{ cm}^{-1}$  due to in-plane vibration well-ordered  $\text{SP}^2$  carbon atoms with a graphitization ratio of ( $I_D/I_G=1.34$ ). Raman spectrum of GO-LDH revealed a band at  $1416\text{ cm}^{-1}$  due to stretching mode of interlayered  $\text{CO}_3^{2-}$  and D- and G-bands were shifted to  $1350\text{ cm}^{-1}$  and  $1608\text{ cm}^{-1}$ , respectively, with ( $I_D/I_G=1.58$ ), indicating the increase in the crystallization degree of LDH by adding GO. Whereas, the  $I_G$  was lower than that of GO due to the perfect formalization of the lamellar structure on GO surface (Rashed et al., 2022). Moreover, CBB adsorption by SDS-LDH depicted that the sharp peak of alkyl chain of SDS exhibited a Raman shift to  $2992\text{ cm}^{-1}$  due to the hydrophobic interaction of alkyl chain of SDS with CBB. Whereas, adsorption of CBB by GO-LDH led to a Raman shift for most peaks as well as D- and G-bands were shifted to  $1373$  and  $1587\text{ cm}^{-1}$ , respectively (Liang et al., 2015), with a decreased graphitization ratio ( $I_D/I_G=1.041$ ) due to the blockage and the reduction of GO-LDH defects by CBB adsorption (Bu et al., 2021). Therefore, Raman spectra confirmed that metal oxides complexation for GO-LDH, CBB/SDS-LDH, and CBB/GO-LDH in the range of  $400-1000\text{ cm}^{-1}$ . The CBB adsorption mechanisms concluded from FTIR and Raman spectra results were represented in the schematic diagrams Fig. S2 (supplementary data).



obtained from SEM (**Danaei et al., 2018**). The zeta potentials of SDS-LDH and GO-LDH were -12 and +16.8 mV, respectively, as displayed in Fig. 7c and d. According to Fig. 7e, the adsorption capacity of SDS-LDH decreased as its initial pH increased with maximum  $q_e$  of 47 mg/g (pH = 1.5 and 3) owing to the increased positive charges on the adsorbent surface that attract the negative charges of CBB dye. The decrease in the adsorption capacity of SDS-LDH above (pH = 7) was due to the increase of OH<sup>-</sup> anions on its surface [1]. However, the maximum adsorption capacity of GO-LDH was 62 mg/g at weak acidic medium (pH = 5.5) as stated for LDH-like materials (**Rashed et al., 2022**), (**Sharif et al., 2020**). The adsorption capacity decreased below and above (pH = 5.5) as a result of GO-LDH metal dissolution in acidic and basic media (**Rashed et al., 2022**), (**Zaghouane-Boudiaf et al., 2012**). From Fig. 7f, the point of zero charge (PZC) of SDS-LDH was 7.6, which was less than that of GO-LDH (7.9) due to the higher negativity of SDS-LDH than GO-LDH. According to the relationship between initial pH (pH<sub>i</sub>) and final pH (pH<sub>f</sub>), Fig. 7g, it was found that below (pH<sub>i</sub> = 7), pH<sub>f</sub> > pH<sub>i</sub> due to the partial protonation of LDH, whereas above (pH<sub>i</sub> = 7), pH<sub>f</sub> was in the range of (7.7 to 8.2) and (7.5 to 8.5) for SDS-LDH and GO-LDH, respectively, due to the buffering adsorption capacity on the adsorbent (**Ai., et al (2011)**).



**Fig. 7:** Zeta-size size distribution of a) SDS-LDH and b) GO-LDH, zeta-potential of c) SDS-LDH, d) GO-LDH, e) adsorption capacity vs pH of SDS-LDH ( $t=60$  min,  $C_0= 40$  ppm, dose= 85 mg,  $T= 20^\circ\text{C}$ ) and GO-LDH ( $t= 10$  min,  $C_0= 40$ , dose= 65 mg, at  $T=20^\circ\text{C}$ ), f) pH difference vs initial pH and g) final pH vs initial pH for SDS-LDH and GO-LDH.

## 3.2. Adsorption, kinetics and isotherm studies

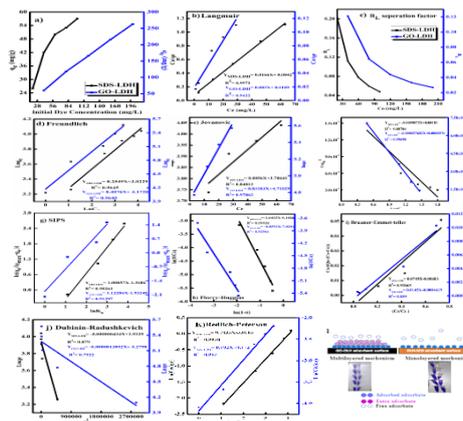
### 3.2.1. Contact time effect and adsorption kinetics

The adsorption capacity of CBB by the synthesized adsorbents varied as the adsorption time proceeded as displayed in Supp Fig. 3. The contact time effect of CBB adsorption was tested at  $20^\circ\text{C}$ , 20 and 40 ppm initial concentration at pH= 7 and 5.5 for 65 mg dose of SDS-LDH and GO-LDH, respectively. The adsorption capacity was so steep to reach 46 and 53 mg/g for the first 10 min and 30 seconds then gradually slowed down to 51 and 62 mg/g till reaching equilibrium at 60 and 5 minutes for SDS-LDH and GO-LDH, respectively. The equilibrium state was due to the super saturation of the CBB on the adsorbents owing to the large interlayered distances of GO-LDH as well as its high surface area, which allowed the rapid diffusion of CBB molecules in a few minutes (**Liang et al., 2015**) as compared to SDS-LDH that relied on the solubility of CBB molecules with SDS alkyl chains “ad-solubilization process” (**Munir et al., 2020**), (**Ramimoghadam, Hussein, & Taufiq-Yap, 2012**). The mechanism of solid-phase adsorption was investigated via various kinetic models as: PFO (**Benjelloun et al. (2021)**), Bhattacharya and Venkobachar, Natarajan and Khalaf (**Srihari & Das, 2008**), PSO, Avrami, Elovich (**Benjelloun et al. (2021)**), Ritchie (**Bayramoğlu et al. (2006)**), Fractional Power (**Srihari & Das, 2008**), Boyd ion exchange model, and intra-particle diffusion (**Ni et al., 2007**), which were expressed by equations as explained in Supp Table 1. The linear regression plots created by the kinetic models with arrangement order PSO > Bhattacharya and Venkobachar > Natarajan and Khalaf > Fractional Power > Elovich > PFO > Avrami > Ritchie; and PSO > Natarajan and Khalaf > Bhattacharya and Venkobachar > Fractional Power > PFO > Elovich > Avrami > Ritchie for SDS-LDH and GO-LDH, respectively as shown in Supp Fig. 3(b-k). In accordance with  $R^2$ , PSO possessed good agreement between the calculated  $q_e$  (51.33 and 63.69 mg/g) and experimental  $q_e$  (51 and 62 mg/g), implying that CBB flawlessly covered SDS-LDH and GO-LDH, respectively (**Rashed et al., 2022**). The initial adsorption rates of SDS-LDH and GO-LDH, were calculated as 45.167 and 311.526 mg g<sup>-1</sup> min<sup>-1</sup>, respectively [50]. Table 1 showed that SDS-LDH obeyed Bhattacharya and Venkobachar with  $K_b$  (0.03912) min<sup>-1</sup>, which well-matched with  $K_1$  (0.0342) min<sup>-1</sup> of PFO than GO-LDH with  $K_n$  (0.5386) min<sup>-1</sup>, which obeyed Natarajan and Khalaf and agreed with  $K_1$  (0.5015) min<sup>-1</sup> of PFO. Furthermore, the adsorbate uptake by the adsorbent positively is correlated with  $\beta$  of

fractional power for SDS-LDH ( $\beta=0.05657$ ) and GO-LDH ( $\beta=0.06886$ ) than that for Elovich. However, Avrami's and Ritchie's data revealed that the adsorption was instantaneous ( $n_{av}<1$ ), obeying diffusive law and ion exchange, the observed low trend of  $R^2$  for Avrami and Ritchie was not applicable for the adsorption system. PSO's kinetic model showed the best fit of all these models and suggested the physisorption nature. The adsorption mechanism was well-defined by Boyd's and the intra-particle diffusion model. Supp Table 1 displayed that Boyd's constant  $\beta=0.039112$  and  $0.50159$  with  $R^2=0.9298$  and  $0.8933$  for SDS-LDH and GO-LDH, respectively, didn't pass the origin. Therefore, not only the film extra-particle diffusion but also intra-particle diffusion was the predominant and the later was the most effective (inner transport > outer transport). The mechanism was demonstrated according to the intra-particle diffusion model that depicted the presence of three linear regions as shown in Supp Fig. 3 (Rashed et al., 2022), (Ni et al., 2007). The first region described the film diffusion, demonstrating the dye diffused from the solution to the external surface of either SDS-LDH or GO-LDH with an increased diffusion constant  $14.577$  and  $74.9647$   $\text{mg/g}\cdot\text{min}^{-1}$  for SDS-LDH and GO-LDH, respectively, along with zero boundary layer thickness for the both adsorbents, as presented in Supp Table 2. The second region described the intra-particle diffusion, referring to dye molecules diffusion via the layers of SDS-LDH and GO-LDH with diffusion constant of  $1.22$  and  $6.275$   $\text{mg/g}\cdot\text{min}^{-1}$ , respectively and then reduced to zero due to the saturation of dye molecules in the third region via intra-particle diffusion, as shown in Supp Table 2. In the second and third region of CBB adsorption, the boundary layer thickness increased till reaching equilibrium of SDS-LDH ( $41.418$  and  $51$   $\text{mg/g}$ ) and GO-LDH ( $47.347$  and  $62$   $\text{mg/g}$ ), respectively, Supp. Table 2. Furthermore, as the boundary layer thickness increased, intra-particle diffusion was not the only rate-determining step (Rashed et al., 2022) as illustrated in Supp Fig. 3

### 3.2.2. Initial dye concentration effect and equilibrium data

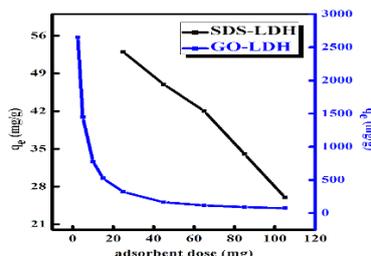
The increase in adsorption capacity curve with increasing initial concentrations of CBB (20-100 and 40-200  $\text{mg/L}$ ) for SDS-LDH (26-57  $\text{mg/g}$ ) and GO-LDH (60-263  $\text{mg/g}$ ), respectively was displayed in Fig. 10a. The adsorption capacity of GO-LDH was vastly larger than that of SDS-LDH as the higher mass driving force of high concentration was directed towards the available high interlayered distance, allowing super CBB adsorption and intercalation (Munir et al., 2020). Demonstratively, SDS-LDH lacked GO-LDH properties, so the hydrophobic interaction with CBB was not effective as the ion-exchange of GO-LDH with CBB. The isothermal behaviour associated with solid-liquid adsorption dynamics was investigated through a comprehensive suite of isotherm models. These encompassed the classical Langmuir and Freundlich models, along with the Harkins-Jura Benjelloun et al. (2021), Jovanovi (Ayawei et al., 2017), Sips, Flory-Huggins, Brunauer-Emmett-Teller, Dubinin-Radushkevich (D-R), and Redlich-Peterson isotherms Benjelloun et al. (2021). An intricate analysis and discussion pertaining to the underlying adsorption mechanisms, as well as the affinity characterizations of the adsorbents, have been delineated in the supplementary information section. The adsorption isotherms plots were displayed in Fig. 8(b-k) and their equilibrium data, based on  $R^2$ , and adsorption parameters were recorded and calculated in Supp Table 3. According to  $R^2$ , the adsorption system lied between the homogenous monolayered and heterogenous multilayered adsorption with arrangement order Langmuir>Redlich-Peterson>Sips>Freundlich>Florry-Huggins>Harkins-Jura>Dubinin-Radushkevich>Bruaner-emmetteller>Jovanovic and Harkins-Jura>Redlich-Peterson>Jovanovic>Freundlich>Langmuir>Bruaner-emmetteller>Florry-Huggins>Sips>Dubinin Radushkevich for SDS-LDH and GO-LDH, respectively. SDS-LDH was observed to perfectly fit Langmuir isotherm with maximum adsorption capacity of ( $q_m=60$   $\text{mg/g}$ ) of homogeneously monolayered coverage with the prove of Redlich-Peterson of ( $\beta=0.7435$   $\text{Lmg}^{-1}$ ) close to unity more than that of GO-LDH ( $q_m=333.33$   $\text{mg/g}$ ,  $\beta=0.59201$   $\text{Lmg}^{-1}$ ). The Langmuir isotherm is eloquently characterized by the dimensionless separation factor, ( $R_L$ ), as illustrated in Figure 8c.  $R_L$  values of the initial concentration of CBB of SDS-LDH ( $0.2016 - 0.0481$   $\text{mg/L}$ ) and GO-LDH ( $0.0646 - 0.0269$   $\text{mg/L}$ ) lied between 0 and 1, referring to strong adsorption due to the low  $R_L$  values (Ai, et al 2011). Moreover, the sips isotherm possessed  $q_{\max}=64.505$   $\text{mg/g}$  with Freundlich adsorption intensity  $1/n=0.255$   $\text{mg/L}$ . The positive Flory-Huggins  $k_d$  indicated that the system was spontaneous and feasible. The harkins-jura, Dubinin Radushkevich, Bruaner-emmetteller, and javonovic isotherms didn't take part in the SDS-LDH adsorption system, based on small  $R^2$ . On the other hand, GO-LDH was found to perfectly fit the Harkins-Jura isotherm ( $A=17336$ ,  $B=1.692$ ), with Redlich Peterson of  $\beta$  of GO-LDH ( $0.59201$   $\text{Lmg}^{-1}$ ), which is decreased due to the high initial concentration of CBB dye obeying Jovanovic isotherm of  $q_{\max}=111.084$   $\text{mg/g}$ , exhibiting mechanical interaction between the adsorbent and the adsorbate, and Freundlich isotherm of adsorption intensity ( $1/n$ ) of  $0.4076$   $\text{mg/L}$ . The Bruaner-emmetteller, flory huggins, sips, and D-R weren't involved in the adsorption system of GO-LDH, based on small  $R^2$ . Hence, it was obvious that SDS-LDH preferred the monolayered adsorption (homogenous nature), while, GO-LDH favored the multilayered adsorption (heterogeneity nature), as discussed in the schematic diagram Fig. 8.



**Fig. 8:** a) Effect of initial concentration of SDS-LDH (PH= 7, t=60 min, dose= 65 mg, T=20 °C) and GO-LDH (PH= 5.5, t= 5 min, dose= 65 mg, T= 20 °C), Adsorption isotherms of b) Langmuir, c)  $R_L$  separation factor, d) Freundlich, e) Harkins-Jura, f) Jovanovic, g) SIPS, h) Florry-Huggins, i) Bruaner-Emmet-teller, j) Dubinin-Radushkevich, k) Redlich-Peterson and l) schematic diagram of multi- and monolayered adsorption mechanism

### 3.2.3. Adsorbent dose effect

The adsorption capacity was investigated by varying the adsorbent dose as shown in Fig. 9. The adsorption capacities of SDS-LDH (53-26 mg/g) and GO-LDH (2646 -73 mg/g) curves were observed to decrease as the adsorbent dose of SDS-LDH (25-105 mg) and GO-LDH (2.5-105 mg) increased. It was found that increasing the dose of either SDS-LDH or GO-LDH adsorbents led to their aggregation, causing the surface area degradation and resulting in low adsorption capacity. However, as the dose of SDS-LDH or GO-LDH decreased, the phases of the adsorbents were easily dispersed, allowing an ease CBB adsorption (Rashed et al., 2022). Illustratively, 5 mg of GO-LDH resulted in an adsorption capacity of 1443 mg/g, which was consistent with results obtained by TMGO (7 mg) for the removal of CBB dye [3].



**Fig. 9:** Effect of adsorbent dose: SDS-LDH (PH= 7, t=60 min,  $C_0 = 40$  ppm, T= 20 °C) and GO-LDH (PH= 5.5, t=5 min,  $C_0 = 80$  ppm, T= 20 °C).

### 3.2.4. Sodium Chloride effect

In the realm of adsorption dynamics, ionic potency is a critical factor that can exhibit an ascending, descending, or steady trend. Evidently, as delineated in Fig. 10, the escalation in sodium chloride concentrations (from 20 to 100 mg) exhibited no discernible alteration in the adsorptive capabilities of SDS-LDH, which steadfastly stood at 41 mg/g. However, a palpable decline in the GO-LDH adsorptive prowess was observed, plummeting from 1240 mg/g to a diminished 870 mg/g. This decrement can be attributed to the repulsive interplay between the negative valences of chloride ions and those of CBB anions, which inevitably curtailed the adsorptive competence of GO-LDH. Contrarily, such electrostatic repulsions bore no influence on the hydrophobic exchanges characterizing the SDS-LDH and CBB liaison (Rashed et al., 2022), (Danaei et al., 2018), (Simonin, 2016)].

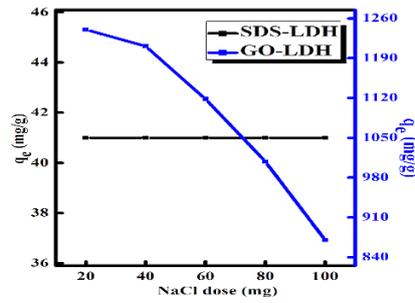


Fig. 10: NaCl dose effect for SDS-LDH (PH=3, t=60 min, C<sub>0</sub>=40 ppm, dose=85 mg, T=20°C) and GO-LDH (PH=5.5, t=5 min, C<sub>0</sub>=80 ppm, dose=5 mg, T=20°C).

### 3.2.5. Temperature effect and thermodynamic studies

The temperature parameter effect on adsorption capacity was displayed in Fig. 11a. As the temperature increased (20°C–80°C), the adsorption capacity reduced for SDS-LDH (47–24 mg/g) and for GO-LDH (1443–1168 mg/g). This may be due to the weakened attractive forces between CBB and SDS-LDH/GO-LDH, allowing the escape of CBB molecules from the solid phase to the liquid phase, thus the adsorption process could be physical adsorption (Abbas et al., 2016). The temperature effect on CBB adsorption by SDS-LDH or GO-LDH was thermodynamically explained to determine the standard enthalpy ( $\Delta H^\circ$ ), standard entropy ( $\Delta S^\circ$ ), and standard free Gibb's energy ( $\Delta G^\circ$ ), using Vant's Hoff equation (Rashed et al., 2022) as displayed in Fig. 11b and Table 4. The entropy was found to possess negative values (Table 4), indicating that the randomness decreased and CBB molecule adsorption on SDS-LDH or GO-LDH was restricted. In addition, negative enthalpy values referred to exothermic adsorption process. Besides, the increase in negativity of Gibb's free energy was corresponding to CBB spontaneous adsorption by either SDS-LDH or GO-LDH (Ibrahim et al., 2019), (Sen et al., 2012)].

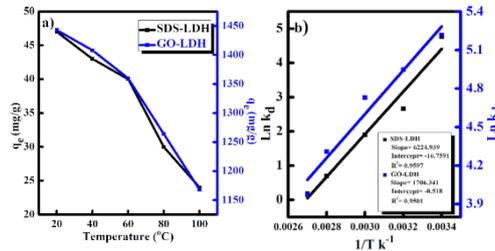


Fig. 11: a) Effect of temperature on SDS-LDH (PH=3, t=60 min, C<sub>0</sub>=40 ppm, dose=85 mg) and GO-LDH (PH 5.5, t= 5 min, C<sub>0</sub> =80 ppm, dose=5 mg) and b) van't hoff plot image.

Table 4: Thermodynamic values for CBB (G-250) dye adsorption onto SDS-LDH and GO-LDH

TEMPERATURE (K)	ENTROPY (J/MOL.K <sup>-1</sup> )	ENTHALPY (KJ/MOL)	FREE GIBBS ENERGY (KJ/MOL)	K <sub>d</sub>
SDS-LDH				
298	-139.3	-51.75	-10.92889	88.8
313	-139.3	-51.75	-5.9456969	22.84
328	-139.3	-51.75	-5.35548	6.92
343	-139.3	-51.75	-2.568773	2.4
358	-139.3	-51.75	0.218	0.932
GO-LDH				
298	-4.306	-14.19	-12.9248	201.49
313	-4.306	-14.19	-12.83867	138.88
328	-4.306	-14.19	-12.75255	100.1
343	-4.306	-14.19	-12.66642	74.88
358	-4.306	-14.19	-12.5803	57.78

### 3.3. Recycling and regeneration studies

LDHs are very important economically in water treatment studies because they can be reused after dye adsorption. Fig. 12a showed the reusability of SDS-LDH and GO-LDH after CBB dye adsorption using the "memory effect" (Starukh et al., 2016), (Starukh & Levytska, 2019), (Mishra, et al., 2018). CBB/SDS-LDH and CBB/GO-LDH were calcined by combustion at 350 °C for 1 hour to remove the adsorbed CBB dye, resulting in calcined double layered hydroxides (C-LDH) and calcined reduced graphene oxide double layered hydroxides (C-RGO-LDH), respectively. The adsorption capacity of C-LDH (47-8 mg/g) and C-RGO-LDH (1443-1320 mg/g) decreased after 5 cycles. Ulibarri et al reported that the reduction in adsorption capacity by C-LDH was due to the elimination of SDS and poor crystallinity caused by calcination, resulting in decreasing dispersion and surface area (Ni et al., 2007). On the contrary, the calcination process had no effect on the properties of C-RGO-LDH that affected the C-LDH. Besides, the high adsorption recycling confirmed that the GO-LDH surface was heterogeneous, obeying the multilayered mechanism and well matched the Harkins-Jura isotherm. The schematic diagrams of Fig. 12b explained the mechanism of regeneration process via memory effect of the tested adsorbents.

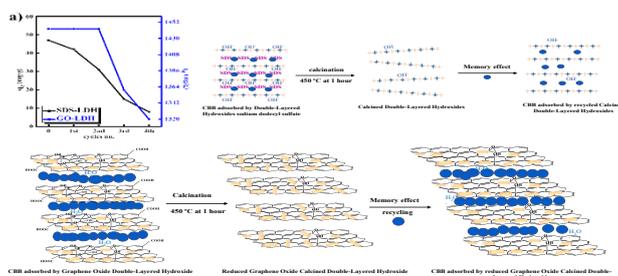


Fig. 12: a) Cycles numbers of regenerated CBB dye adsorption by SDS-LDH (PH= 3, t= 60 min, C<sub>0</sub>= 40 ppm dose= 65 mg, T= 20°C) and GO-LDH (PH= 5.5, t= 5 min, C<sub>0</sub>= 80 ppm, dose=5 mg, T= 20°C) and Schematic diagrams of memory effect of b) SDS-LDH and c) GO-LDH.

## 4. Conclusion

In this context the synthesis of SDS-LDH and GO-LDH via the reconstruction method and the sonochemical method, respectively, for the elimination of CBB organic dye pollutants from an aqueous solution was studied. The morphology of SDS-LDH and GO-LDH were shown to be lamellar structured with a specific average surface area of  $35.5 \times 10^6 \text{ nm}^2/\text{g}$  and  $47 \times 10^6 \text{ nm}^2/\text{g}$ , as well as polydispersity index of 0.34 and 0.406, and an interlamellar distance of 8.8 Å and 11.043 Å, respectively, proving the high element purity of Zn, Al, C, O, and Na. CBB adsorption mechanisms of SDS-LDH and GO-LDH were via hydrophobic interaction and ion-exchange, respectively, inducing better thermal stability of CBB by GO-LDH than SDS-LDH with weight loss of 0.269 mg and 0.699 mg, respectively. Accordingly, at room temperature, the q<sub>e</sub> of GO-LDH (1443 mg/g) at (pH = 5.5, t= 5 mins, C<sub>0</sub>= 80 ppm, dose = 5 mg) was higher than that of SDS-LDH (62 mg/g) at (pH = 3, t= 60 mins, C<sub>0</sub>= 40 ppm, dose = 85 mg), following the PSO kinetic model with initial velocity  $45.167 \text{ mg g}^{-1} \text{ min}^{-1}$  and  $311.526 \text{ mg g}^{-1} \text{ min}^{-1}$  for SDS-LDH and GO-LDH, respectively and controlled by the film and intra-particle diffusion mechanism. Additionally, SDS-LDH and GO-LDH follow the monolayered mechanism of the Langmuir isotherm and the multilayered mechanism of the Harkins-Jura isotherm, respectively. The CBB adsorption process was restricted ( $-\Delta S^\circ$ ), exothermic ( $-\Delta H^\circ$ ), spontaneous ( $-\Delta G^\circ$ ), and physical. Using the memory effect of the adsorbents, C-LDH and RGO-C-LDH were recycled, showing that RGO-C-LDH (1443-1320 mg/g) was more efficient than that of C-LDH (47-8 mg/g) after five runs. These results show that GO-LDH was sustainable adsorbent because of its nearly zero-cost production in the wastewater industry.

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