

Zinc-aluminium layered double hydroxides: Fabrication, study and adsorption application for removal organic dye from aqueous media

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Abstract

The current research aimed for the preparation of the zinc-aluminium layered double hydroxide by co-precipitation method using metal nitrates and sodium hydroxide. The obtained product was studied using X-ray diffraction, High-resolution transmission electron microscopy, Fourier transform infrared spectroscopy and Field-emission scanning electron microscopy. XRD data appeared that the obtained zinc-aluminium layered double hydroxide is in the range of the nanoparticle. The crystallite and particle sizes were determined from XRD and TEM. The adsorption parameters of organic dye removal using the zinc-aluminium layered double hydroxides were investigated using a batch method. The adsorption isotherms and kinetic models were studied utilizing the synthesized zinc-aluminium layered double hydroxide.

Keywords: Co-precipitation method, Zn-Al layered double hydroxide, batch method, Morphology, organic dyes.

1. Introduction

Water is one of renewable and fundamental natural resources which is essential for survival all forms of life and human being. Water is the most appropriate solvent which easily dissolves other substances. Pure water is primarily used in the drinking, food, cooking, industrial activities, navigation, agriculture etc. The contamination of water is happened when some substances are found to such a degree that water is unable to be used in different purpose[1, 2]. The pollutants of water are categorized into the soluble and insoluble of organic and inorganic materials. Inorganic and organic pollutants have different examples such as heavy metals, herbicides, insecticides, dyes, fertilizers from agricultural, phosphate, nitrate, food processing wastes, pharmaceutical waste, volatile organic substances, and chemicals, etc. Organic pigments are the major source of environmental of water from textile, paper, plastic, drug, and other industries[3-5].

Dyes are considered to have higher stability and are complex to biodegrade because they generally are not easy synthetic and complicated structures. Dyes are separated from contaminated water by using different advanced techniques [2, 6, 7] like adsorption, oxidation, catalytic photodegradation, coagulation, etc. The technique of adsorption is a highly effective separation method, which is preferable to water treatment of other technologies. The main advantages of this technique are low cost, simplicity of setup, design, ease of operation and sensitivity to different poisonous substances. Sunset dye is an example of the common types of anionic dye for coloring beverages and a variety of foods including confectionary, desserts, soups, cheeses, savory snacks, and preserved fruits. Contaminated water acts as a danger to human and it can't be used in industrial field. Sunset dye can produce some dangerous outcome in humans such as provoke allergic reaction which can be exacerbated in people who are already asthma or urticarial sufferers. The extraction of sunset dye is very important to preserve the water from contamination.

Various nanomaterials in the form of oxide, sulphide, hydroxide, layered double hydroxides and

other nanoparticles[8-10], which used in various applications such as catalyst, adsorbent, pigment, ceramic, etc. The term of layered double hydroxides (LDH) is defined as inorganic materials with anion intercalated functional group, it also known as anionic clays or hydrotalcite compounds. In LDH, the metal hydroxide has a positive charge by which host layers stack alternately with interlayer anions result in a sandwich structure. The metal ions positioned in the host layer coordinated divalent M(II) and trivalent M(III) cations octahedrally and the anions in the interlayer interact with the metal ion host layer by using various types of bonds such as hydrogen bonding, ionic bonding, and Vander Waals forces[11-14]. In the present study, Zn-Al layered double hydroxides (LDH) have been synthesized using the coprecipitation method, followed by the characterization by the different tools. The fabricated Zn-Al layered double hydroxides (LDH) are used as candidate adsorbent for sunset dye using the batch method.

2. Experimental

2.1. Materials and Reagents

All chemicals used in this work were purchased and used as received without any further purification. Aluminum nitrate nonahydrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 98 %) were obtained from Alpha Chemika Chemical Company, India. Zinc nitrate ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, 98%) was purchased from Qualikems Chemical Company, India. Sunset yellow dye ($\text{C}_{16}\text{H}_{10}\text{N}_2\text{Na}_2\text{O}_7\text{S}_2$, 95%; Molecular Weight: 452.37 g/mole; $\lambda_{\text{max}} = 480 \text{ nm}$) were obtained from Sigma-Aldrich Chemical Company. Nitric acid (HNO_3 , 69%), Hydrochloric acid (HCl , 37%) and sodium hydroxide (NaOH , 99%) were purchased from El Nasr pharmaceutical chemical company. All chemicals and reagents were of analytical grade and used as received without any purification. Freshly bidistilled water was used through all experiments. The chemical structure of the sunset dye is shown in Fig. 1.

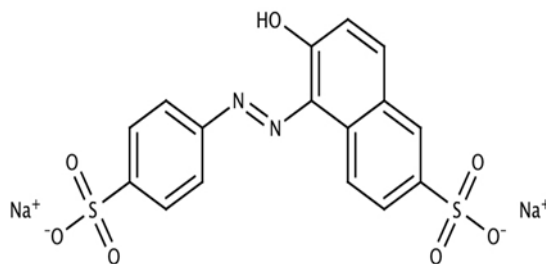


Fig. (1) The chemical structure of sunset dye.

2.2. Preparation of Zn-Al layered double hydroxides

Zinc aluminum layered double hydroxides (Z-A LDH) were prepared by the co-precipitation method. 3.7513 gram of aluminum nitrate nonahydrate was dissolved in 75 ml bidistilled water. Zinc nitrate hexahydrate solution (0.03mole) was added to the pervarsity solution under the stirring at room temperature. The obtained Zn-Al solution was adjusted at pH=9 by the addition of 15 ml of 3 M of NaOH with vigorous magnetic stirring for 15 min. The fabricated precipitate was ultrasound using probe ultrasonication for 20 min with continuous stirring, followed by the standing for 24 h. Then, the formed precipitate was separated by the centrifugation for 5 min and washed four times with deionized water. The separated precipitate was dried at 50 °C for 12 h. The obtained sample were named as Z1, related to Zn/Al molar ratio = 3:1.

2.3. Characterization tools

The obtained Z1 sample was recorded using an FTIR spectrometer (FTIR: Jasco FTIR 460 plus) at room temperature from 4000 to 400 cm^{-1} . The appeared phase of the synthesized Z1 sample was investigated from the XRD patterns using 18 KW diffractometer (Bruker; model D8 advance) with monochromatic Cu-K α radiation, 1.54178 (Å) in the angular range of 5-80° with step size 0.02° (2 θ) and scan step time 0.4 (s). The adsorption process was investigated using a Jasco UV-Vis spectrophotometer (Jasco; model V 670). The morphology of the synthesized Z1 sample was

investigated using FE-SEM (FE-SEM, Zeiss Sigma 500 VP analytical FE-SEM, Carl Zeiss, Germany). The morphology of the synthesized Z1 sample was tested HR-TEM (HR-TEM: JEOL; model, 200 Ex and an electron voltage of 200 KV). A small amount of the materials was dispersed in 10 mL water then sonicated for 30 minutes. Few drops of the resulting suspension were placed on a covered copper coated carbon grid.

3. Results and Discussion

3.1. Characterization of Zn-Al layered double hydroxides

Zinc aluminum layered double hydroxides were characterized using X-ray diffraction as shown in Figure 2. Due to the XRD pattern of the zinc aluminum layered double hydroxides, the presence of the sharp diffraction peaks in their diffraction patterns reflected the presence the crystalline of zinc and aluminium hydroxides. From XRD pattern, the peaks of the synthesized Z1 sample were found at $2\theta=10.047, 10.557, 11.871, 20.152, 23.731, 29.611, 32.127, 34.848, 39.487, 47.022, 60.341$ and 61.836 , indicating that the crystalline zinc aluminum layered double hydroxides obtained using the coprecipitation method. The crystallite size (D , nm) of the Z-A LDH determined using the Scherrer equation ($D = 0.9\lambda/\beta\cos\theta_B$). Where λ is the wavelength of X-ray radiation, D is constant, β is the full width at half maximum of the diffraction peak and θ_B is the Bragg diffraction angle. The crystallite size determined from XRD pattern to be 29 nm.

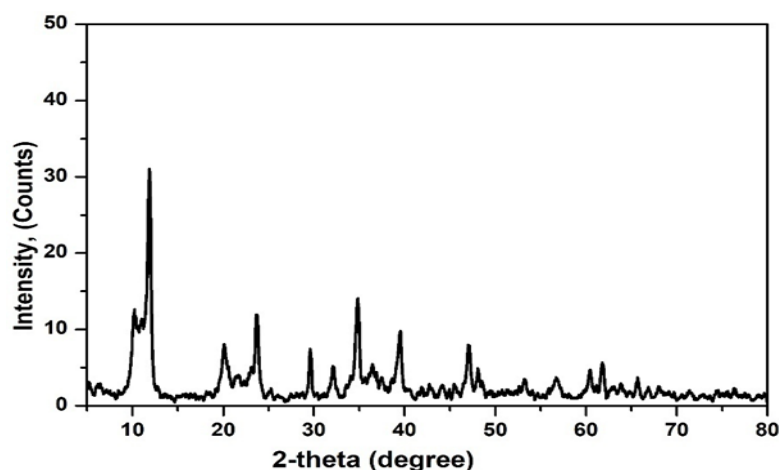


Fig. (2) XRD of zinc aluminum layered double hydroxides (Z1 sample) using coprecipitation method.

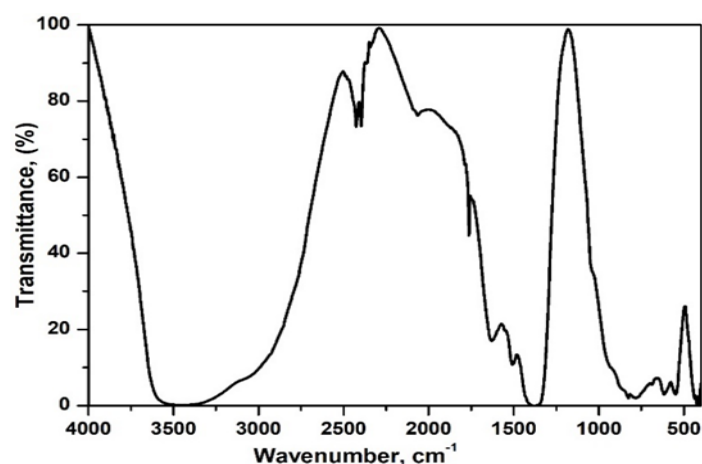


Fig. (3) FTIR of zinc aluminum layered double hydroxides (Z1 sample) using coprecipitation method.

The FTIR spectra of the synthesized Z1 sample appeared the characteristic bonds corresponding to zinc aluminum layered double hydroxides as represented in Figure 3. The broad and strong peak related in the range of 300-3600 cm^{-1} and 1620 cm^{-1} is attributed to the stretching and bending modes of hydroxyl groups of water molecules on the external surface and inside the interlayer position zinc aluminum layered double hydroxides. The strong peak at 1390 cm^{-1} is related the

vibration mode of the NO_3^- interlayer anions in the synthesized zinc aluminum layered double hydroxides. Also, the peaks in the range of 400-1100 (420cm^{-1} , 550cm^{-1} , 620cm^{-1} , 790cm^{-1} , 820cm^{-1} and shoulder at 950 and 1050cm^{-1}), are related to the tetrahedral and octahedral positions of the Zn-O, Al-O, O-Zn-O and O-Al-O inside the fabricated zinc aluminum layered double hydroxides[15].

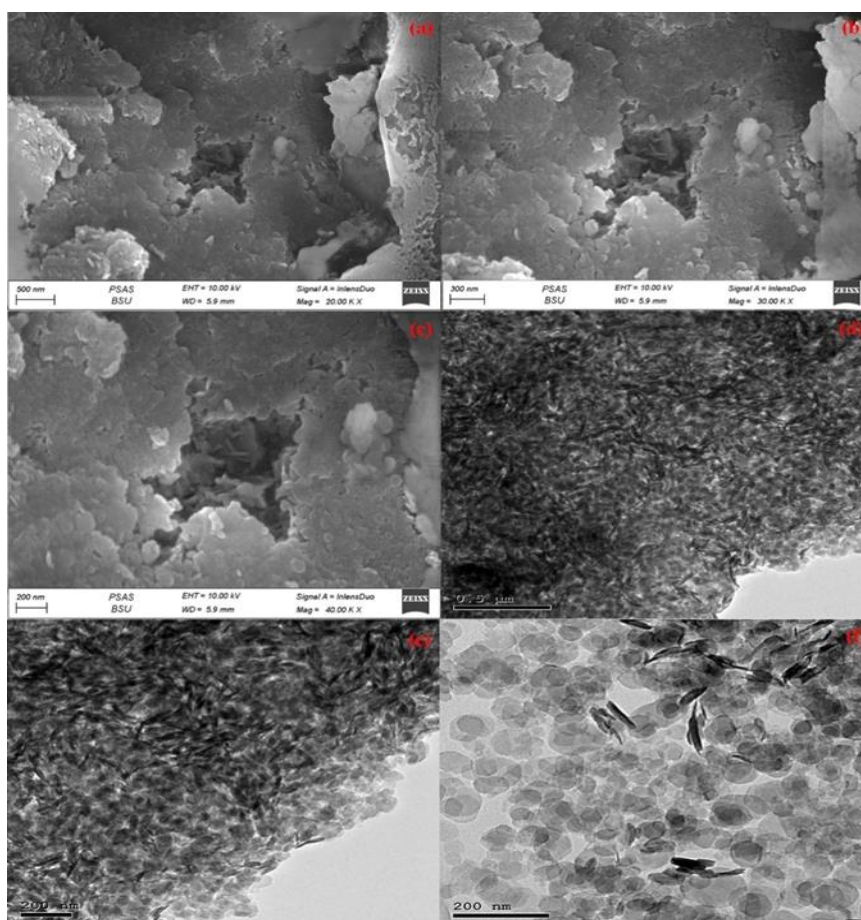


Fig. (4) HR-TEM and FE-SEM of zinc aluminum layered double hydroxides (Z1 sample).

The morphology, particle and grain sizes of the fabricated zinc aluminum layered double hydroxides (Z1 sample) were investigated using TEM and SEM as shown in Figure 4. TEM micrographs display the presence of spherical shaped and particle size determined to be 44 nm. SEM images appeared the complete and incomplete spherical shaped in the form of sheets and platelets. Grain sizes calculated from the SEM micrographs to be 80 nm. According to the obtained particle and grain sizes, the synthesized zinc aluminum layered double hydroxides (Z1 sample) is formed with hard agglomeration comparison with the crystalline size.

3.2. Adsorption studies

The obtained Z1 sample were used as adsorbents for the rapid removal of sunset yellow dye using batch mode. Adsorption parameters of the removal of sunset yellow dye using synthesized zinc aluminum layered double hydroxides (Z1 sample) were tested by the study of the different adsorption conditions such as pH, contact time, adsorbent dosage, and initial dye concentrations. The values of removal percentage were calculated using the measured and initial dye concentrations of sunset yellow dye over the synthesized zinc aluminum layered double hydroxides as adsorbent. From batch mode, the extracted data were used to quantify the adsorption capacity (Q_e) and the removal efficiency (R %) [16] from equation No.1 and 2, respectively.

$$Q_e = \frac{(F_o - F_t)V}{m} \quad (1)$$

$$\text{Removal (R, \%)} = \frac{(F_o - F_t)}{F_o} \times 100 \quad (2)$$

Where, Q_e is the uptake of dye at certain time (mg g^{-1}), V is the volume of solution (L), F_o is the initial dye concentration (mg/L), F_t is the dye concentration at time t (mg/L), and m is the weight (dosage of the sorbent) of the adsorbent (g).

The effect of initial pH was studied with various pH values (2 – 9) using the synthesized Z1 sample as displayed in Figure 5(a). In the experiment, 50 mg of Z1

sample was added to 25 mL sunset yellow dye aqueous solution with an initial concentration of 100 mg/L. The efficiency of dye removal was estimated by measuring the remaining dye after 2 h at $\lambda_{\text{max}} = 482 \text{ nm}$ for sunset dye. The removal efficiency recorded high values above 97.8-99.8 % for all pH ranges. The pH 7 selected as neutral pH with removal value (99.8 %).

The contact time of the removal 100 mg/L of sunset was tested using 50 mg of Z1 sample at pH= 7 and the equilibrium time was attained in the ranging between 1 and 35 min as shown in Figure 5(b). The rate of uptake was rapid in the beginning (0-5 min) and became gradually in the latter stages till reached saturation state (5-35 min). The maximum separation and removal of sunset yellow dye was attained in 30 min to be 48 mg/g.

The effect of the quantity of Z1 sample on the removal 100 mg/L of sunset yellow dye was investigated by adding various amount (0.01- 0.1 g) of Z1 sample as adsorbent and the pH of the solutions were fixed at 7. After 30 min, the efficiencies were calculated as shown in Figure 5(c). The extracted data indicated that the increasing of adsorbent dosage leads to the increase of the separation of sunset yellow dye on Z1 sample due to the rising of the active center on the surface of zinc aluminum layered double hydroxides (Z1 sample).

Figure 5(d) displays the effect of various initial dye concentration (50-300 mg/mL) for the separation of sunset yellow dye over 0.05 g of Z1 sample adjusting to pH = 7. After 30 min, the adsorption capacities were determined from the extracted data. The rate of uptake was rapid in the beginning and reached saturation because of the maximum availability of active sites on the surface of Z1 sample. As the time passed, the active sites were blocked, the rate of the separation of dye decreased. The maximum capacity of zinc aluminum layered double hydroxides calculated to be 94 mg/g for removal of sunset yellow dye after 30 min.

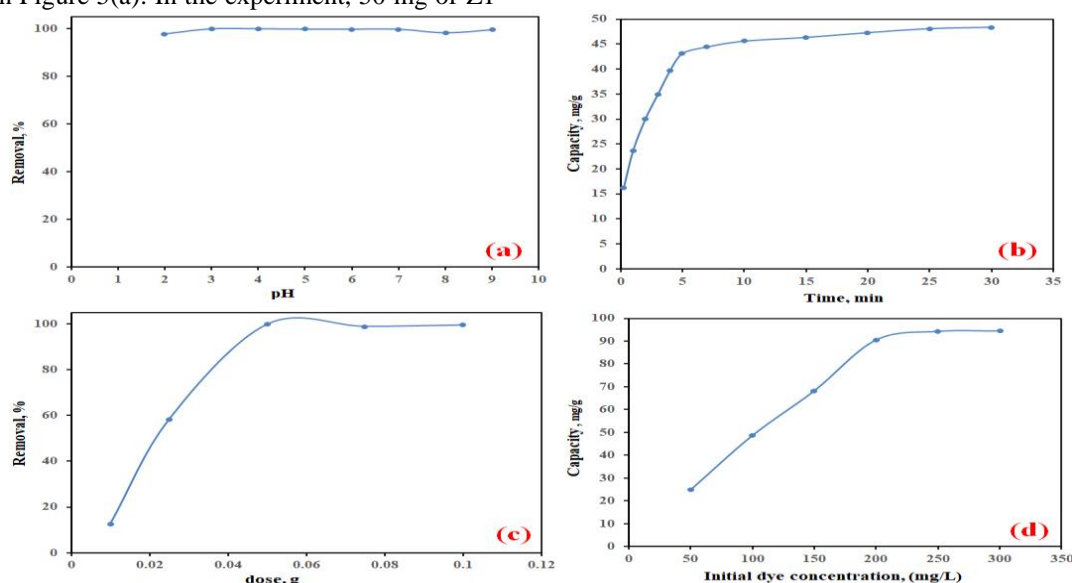


Fig. (5) Effect of pH (a), time (b), adsorbent dose (c) and initial concentration on the removal of the sunset yellow dye using the zinc aluminum layered double hydroxides (Z1 sample).

3.3. Adsorption isotherms studies

The adsorption capacities with the various initial concentrations of the sunset yellow dye (50 and 300 mg/L) for Z1 sample were studied by the adsorption models using Langmuir and Freundlich isotherms. The experimental data are used for the description of adsorption behavior of sunset yellow dye on 0.05 gram of the synthesized Z1 sample at pH=7 and 293 K. The equations of Langmuir, Freundlich and Temkin [16-20] are displayed in No. 3, and 5, respectively.

$$\frac{F_e}{Q_e} = \frac{1}{K_L Q_m} + \frac{F_e}{Q_m} \quad (3)$$

$$\ln F_e = \ln K_F + \frac{1}{n} \ln F_e \quad (4)$$

$$Q_e = Z \ln K_T + Z \ln F_e \quad (5)$$

Where, F_e is the equilibrium concentration of sunset yellow dye in solution (mg/L), Q_e is the equilibrium adsorption capacity of sunset yellow dye on the adsorbent, K_L is the Langmuir parameter (L/mg), q_m is the maximum amount of adsorbed sunset yellow dye to adsorbent (mg/g), K_F is the Freundlich constant (mg/g) and $(1/n)$ is the heterogeneity factor. K_T is the equilibrium binding constant (L/mol) corresponding to the maximum binding energy and constant Y is related to the heat of adsorption. The slope and intercept used for the determination of the constants of Langmuir, Freundlich and Temkin isotherms from a plot of $[F_e/Q_e \text{ versus } F_e]$, $[\ln Q_e \text{ versus } \ln F_e]$, and $[q_e \text{ versus } \ln F_e]$, respectively as shown in Figure 6(a-c).

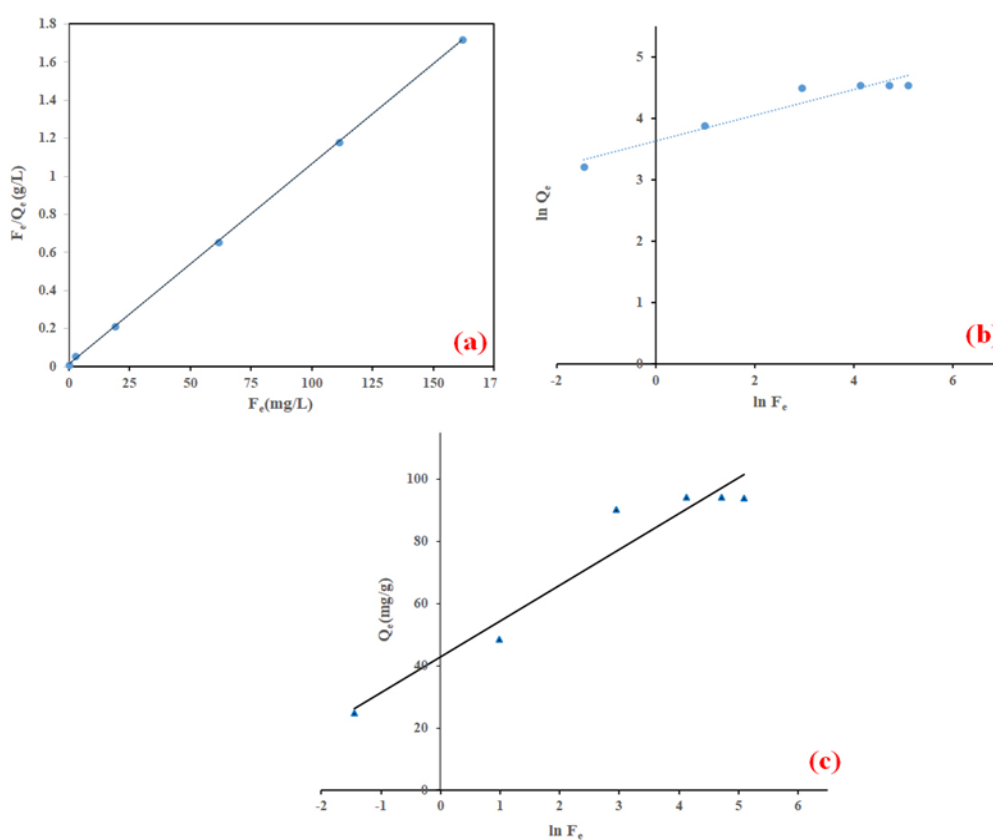


Fig. (6) Langmuir (a), Freundlich (b) and Temkin (c) models for removal the sunset yellow dye on zinc aluminum layered double hydroxides (Z1 sample).

From the R^2 values, the Langmuir model ($R^2=0.999$) of the sunset yellow dye adsorption on the Z1 sample is better fitting than the other models. It means that the removal of the sunset yellow dye on the zinc aluminum layered double hydroxides as adsorbent (Z1 sample) is of monolayer without any interaction between sunset yellow molecules. The values of adsorption capacity: 95.25 mg/g and 93.00 mg/g are the theoretical and experimental values from Langmuir model, respectively. Table 1 is summarized the values were calculated from Langmuir, Temkin and Freundlich models. R_L values were calculated to be 0.00678 - 0.03934, related to the initial concentration 50-300 mg/g. It explains the adsorption process of the sunset yellow dye on the Z1 sample is a favorable adsorption process. Due to the b value from Temkin model, the heat of adsorption (Y) calculated to be 0.1987 KJ/mole and it reflected that the weak interaction force between the sunset yellow dye on the Z1 sample.

Table (1) The extracted parameters from Langmuir, Freundlich and Temkin models for removal of sunset yellow using zinc aluminum layered double hydroxides adsorbent (Z1 sample)

Adsorption isotherm	Parameter	Values
		ZGA
Langmuir	K_L (L/mg)	0.4884
	q_m (cal) (mg/g)	95.24
	R_L	0.00678-0.03934
	R^2	0.999
Freundlich	q_m (exp) (mg/g)	93.00
	K_F [(L/mg) (L/mg) ^{1/n}]	40.10
	q_m (cal) (mg/g)	113.25
	n (L/mg)	4.41
	R^2	0.927
Temkin	q_m (exp) (mg/g)	93.00
	K_T (L/mg)	32.16
	Y (J/mol)	198.7
	Z	3.9175
	R^2	0.935

3.4. Adsorption kinetics

The study of adsorption kinetics is used for the description of the rate of the removal of sunset yellow dye on the synthesized zinc aluminum layered double hydroxides from aqueous solutions. Also, it used for the understanding of the mechanism of adsorption process. The adsorption capacity of zinc aluminum layered double hydroxides sample increased with time and reached to maximum as shown in Figure 7(a, b and c). Kinetic factors of the removal sunset yellow dye on Z1 sample were detected using pseudo-first order, pseudo-second-order equations and intraparticle diffusion model [16-20] as shown in the following equations No. 6, 7 and 8. The kinetic parameters calculated from relations and summarized in Table 1.

$$\log(Q_e - Q_t) = \log(Q_e) - \frac{K_F}{2.303} t \quad (6)$$

$$\frac{t}{Q_t} = \frac{1}{K_S Q_e^2} + \frac{t}{Q_e} \quad (7)$$

$$Q_t = K_{ID} t^{0.5} + E \quad (8)$$

Where, Q_e and Q_t are the amounts of dye adsorbed (mg/g) at equilibrium and time t (min), respectively. t (min) is contact time, and k_F is pseudo-first-order rate constant of adsorption (min^{-1}). K_S (g/mg. min) is pseudo-second-order rate adsorption constant. K_{ID} is intraparticle diffusion constant ($\text{mg/g. min}^{0.5}$), and E is the thickness of boundary layer (mg/g).

According to the plot of $[t/q_t$ against time $t]$, $[\log (q_e - q_t)$ against $t]$ and $[qt$ vs. square root of time] for pseudo-second-order model, pseudo-first-order model and intraparticle diffusion as displayed in Figure 7(a, b and c). The data extracted from Figure 7(a, b and c) and summarized in Table 2. From the R^2 values, the pseudo-second-order ($R^2=0.996$) is the better fitting for the removal of sunset yellow dye on the zinc aluminum layered double hydroxides as adsorbent, than the pseudo first order ($R^2=0.979$). The theoretical and experimental adsorption capacities detected for the adsorption of sunset yellow dye on the zinc aluminum layered double hydroxides, to be 50.74 mg/g and 48.7 mg/g, respectively.

If intra-particle diffusion can explain the mechanism of the removal of sunset yellow dye on the zinc aluminum layered double hydroxides as adsorbent, a straight line must be passed through the origin with $E = 0$. Table (2) appeared the detected values for K_{ID} and E and it displayed that the plot does not pass the origin. It is reflected that the intraparticle diffusion is not the only model for the description mechanism of the adsorption of the sunset yellow dye on the zinc aluminum layered double hydroxides. The adsorption process may be explained by the other types of mechanisms including bulk diffusion, etc.

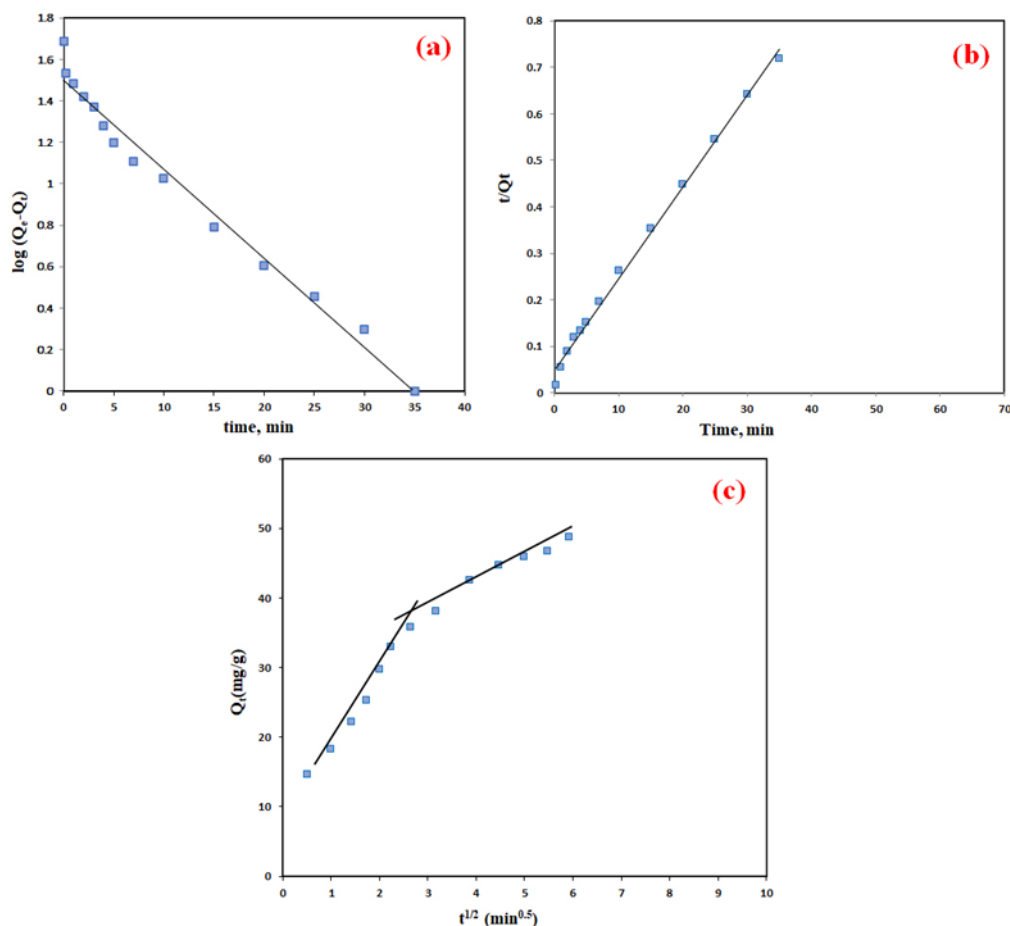


Fig. (7) Pseudo-first-order(a), pseudo-second-order(b) and intraparticle diffusion (c)models for the removal of sunset yellow dye on the zinc aluminum layered double hydroxides (Z1 sample).

Table (2) Adsorption kinetic values of the removal of sunset yellow dye on the zinc aluminum layered double hydroxides (Z1 sample)

Kinetic parameters	Parameter	Values
Pseudo first order	$K_1(\text{min}^{-1})$	0.043
	$q_m(\text{cal})(\text{mg/g})$	31.7
	R^2	0.979
	$q_m(\text{exp})(\text{mg/g})$	48.7
Pseudo second order	$K_2(\text{g/mg}\cdot\text{min})$	0.00799
	$q_m(\text{cal})(\text{mg/g})$	50.74
	R^2	0.996
	$q_m(\text{exp})(\text{mg/g})$	48.7
Intra-particle diffusion	(I) $K_i(\text{mg/g}\cdot\text{min}^{0.5})$	10.458
	$C(\text{mg/g})$	8.39
	R^2	0.986
	(II) $K_i(\text{mg/g}\cdot\text{min}^{0.5})$	3.55
	$E(\text{mg/g})$	27.92
	R^2	0.955

Conclusions

In this research, layered double hydroxides have been successfully fabricated as an adsorbent to the adsorption of the sunset dye from the wastewater. The obtained Z1 sample were characterized by employing different tools. The crystallite, particle and grain sizes were determined from XRD, TEM and SEM to be 29 nm, 44 nm and 80 nm, respectively. The adsorption parameters of the removal of sunset yellow dye using the zinc-aluminium layered double hydroxides were determined using a batch method. The extracted data from isotherm models appeared that Langmuir adsorption isotherm was well fitted than the other models. The pseudo second order ($R^2=0.996$) is the better fitting than the pseudo first order ($R^2=0.979$) for the adsorption of sunset yellow dye on the zinc aluminum layered double hydroxides (Z1 sample). The theoretical and experimental adsorption capacities detected for the adsorption of sunset yellow dye on the zinc aluminum layered double hydroxides, to be 95.25 mg/g and 93.00 mg/g, respectively.

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