

การเตรียม CoAl-เลเยอร์ดับเบิลไฮดรอกไซด์ที่ถูกปรับปรุงพื้นผิว ด้วยโดเดคซิลซัลเฟต และการกำจัดสีย้อม

Preparation of Dodecylsulfate Modified CoAl-Layered Double Hydroxide and Dye Removal

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บทคัดย่อ

งานวิจัยนี้ได้ปรับปรุงสมบัติของตัวดูดซับ CoAl-LDH โดยการอินเทอร์คาเลชันด้วยโดเดคซิลซัลเฟต (DS⁻) ไอออน โดยการทำปฏิกิริยาระหว่างสารแขวนลอย CoAl-LDH ในน้ำ และสารละลายโซเดียมโดเดคซิลซัลเฟต ด้วยวิธีไฮโดรเทอร์มอล ที่อุณหภูมิ 120 °C เป็นเวลา 1 วัน และปรับเปลี่ยนปริมาณ DS⁻ เป็น 5, 10, 20 และ 30 เท่าของความจุในการแลกเปลี่ยนไอออนลบ (AEC) ของ LDH จากนั้นทำการพิสูจน์เอกลักษณ์ของตัวดูดซับอนินทรีย์ที่เตรียมได้ด้วยเทคนิคการเลี้ยวเบนของรังสีเอกซ์ (XRD) และฟูเรียร์ทรานสฟอร์มอินฟราเรดสเปกโทรสโกปี (FTIR) จากรูปแบบการเลี้ยวเบนของรังสีเอกซ์ พบว่า ช่องว่างระหว่างเลเยอร์ของ CoAl-LDH ขยายกว้างขึ้นตามการเพิ่มปริมาณ DS⁻ จนถึง 30 เท่าของ AEC ซึ่งยืนยันการมีโมเลกุล DS⁻ ในช่องว่างระหว่างเลเยอร์ที่จัดเรียงตัวแบบ paraffin monolayer นอกจากนี้ได้มีการศึกษาสมบัติการดูดซับสารละลายสีย้อมโรดามีน 6 จี ออร์เรนจ์ II และอะลิซาริน พบว่า ตัวดูดซับ CoAl-LDH ที่ถูกปรับปรุงพื้นผิวด้วย DS⁻ (30 เท่าของ AEC) ดูดซับโมเลกุลสีย้อมทั้ง 3 สปีชีส์ ได้ดีกว่าตัวดูดซับอื่นๆ และให้ประสิทธิภาพการกำจัดสีย้อมออร์เรนจ์ II สูงที่สุด

คำสำคัญ: CoAl-เลเยอร์ดับเบิลไฮดรอกไซด์, ตัวดูดซับ, สีย้อม, โดเดคซิลซัลเฟต

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ABSTRACT

In this work, the modification of CoAl-LDH adsorbent was investigated by the intercalation of a series of dodecylsulfate (DS^-) ions. The reaction between an aqueous suspension of CoAl-LDH and an aqueous solution of sodium dodecylsulfate was conducted under hydrothermal method at 120°C for 1 day. The adding amount of DS^- was adjusted to be 5, 10, 20 and 30 times to anion exchange capacity (AEC) of the LDH. The as-prepared inorganic adsorbents were characterized by powder X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR). The XRD patterns revealed that the interlayer space of CoAl-LDH was slightly expanded with increasing the loading amount of DS^- until 30 times of the AEC. This result confirmed the presence of DS^- molecules in the interlayer space with paraffin monolayer arrangement. The property of the adsorbents was assessed by the removal of rhodamine 6G, orange II and alizarin from solution. CoAl-LDH modified with dodecylsulfate (30 times of AEC) adsorbed three species of dye molecules from solution more than the other adsorbents, and exhibited the highest efficiency for removal of orange II.

Key words: CoAl-layered double hydroxide, adsorbent, dye, dodecylsulfate

INTRODUCTION

Dye, an organic pigment, is well known for its negative effect on health and water environment. The contamination of dyes in water resource is one of the big issues because the dye molecules possess non-biodegradable and physicochemically stable structure, as well as, bioaccumulation (Ahmad *et al.*, 2015). The production and corresponding discharge of dyes by concerning textile industry are undeniably increased as marketing demand as a result of water pollution. However, purification activities show high removal efficiency such as chemical oxidation/reduction using photocatalyst, ozonation, reverse osmosis, electro-dialysis and so forth, these strategies are sometimes unavailable on procedures due to expensive and complex operations. In comparison with all above methods, adsorption technique is capacitated for cleaning wastewater from overall chemicals (heavy metal, pesticide, organics, etc.) as economically and easy processes (Zhang *et al.*, 2014; Kausar *et al.*, 2018). Therefore, the synthesis and development of potential adsorbent are needed to study for enhancing efficiency for removal of pollutants contaminated in water resources. During the past decades, the preparation of inorganic materials and their applications received much attention

due to the robust structure, large surface area, and recyclability (Kausar *et al.*, 2018). One of them, 2D-layered inorganic adsorbent shows additional advantages such as ion exchangeable capacity, feasible interlayer space and swelling property (Zhang *et al.*, 2014; Sun *et al.*, 2015; Yang *et al.*, 2019).

Layered double hydroxide (LDH) is a candidate of host and/or support materials, which is widely attracted by a number of researchers for applying as catalyst, adsorbent, drug carrier and so on (Liu *et al.*, 2006; Yang *et al.*, 2019). Besides, positive charge on metal hydroxide nanosheets is also a dominant choice from other materials. The typical structure of LDH, $[\text{M}^{3+}_y\text{M}^{2+}_{1-y}(\text{OH})_2]^{y+}(\text{A}^{n-})_{y/n}\cdot z\text{H}_2\text{O}$, comprised of coordinated MO_6 octahedron formed by isomorphic substitution of divalent (M^{2+}) cation in some sites of brucite-like structure by trivalent (M^{3+}) one and sharing edges by oxygen, and water, as well as anionic guest (A^{n-}) species for satisfying charge neutrality in the interlayer space (Yang *et al.*, 2019).

The use of LDH as adsorbent exhibited efficient removal of the negative-charged dye as a result of binding with electrostatic interaction. Nevertheless, dye molecules contaminated in wastewater

compose of various species and especially different charge (positive, negative and/or neutral) that is limited for only reactionary-charged adsorbent. The adsorbent must be modified so that the application is multifunctional and available for all species of dyes. In the concept, the compensation of the positively charged surface of the LDH with negative surfactant makes the adsorbent versatile on different charge-dye molecules. Kong *et al.* (2018) demonstrated that the inorganic material modified with dodecylsulfate could bind the dye molecules with hydrophobic and hydrogen bonding interactions. The intercalation of surfactants in the interlayer space of clay host has been investigated so far and the adsorption of organic guests is successful (Khaorapapong *et al.*, 2011; Demel *et al.*, 2014; Ogawa and Hiramane, 2014). As potential adsorbent on removal of the dye molecules, in this study, the modification of CoAl-LDH was investigated by incorporation of dodecylsulfate in the interlayer space under hydrothermal reaction, and the difference in the adding amounts of dodecylsulfate was also adjusted. The organo CoAl-LDH was evaluated by the adsorption efficiency of rhodamine 6G, orange II and alizarin molecules in solution.

MATERIALS AND METHOD

Materials

Chloride salts of cobalt (CoCl_2) and aluminium ($\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$) were purchased from Carlo Erba Reagenti. Urea ($(\text{NH}_2)_2\text{CO}$) was purchased from Asia Pacific Specialty Chemicals LTD. Sodium dodecylsulfate ($\text{NaC}_{12}\text{H}_{25}\text{SO}_4$) was purchased from Ajax Finechem Pty LTD. Rhodamine 6G ($\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$), orange II ($\text{C}_{16}\text{H}_{11}\text{N}_2\text{NaO}_4\text{S}$) and alizarin ($\text{C}_{14}\text{H}_8\text{O}_4$) were purchased from Acros. All chemicals are the reagent grade and were used directly without any further purification.

Preparation of CoAl-LDH and $\text{DS}_x/\text{CoAl-LDH}$

The synthesis of CoAl-LDH was conducted by the reaction in a two-neck round-bottom flask that equipped with a reflux condition. The aqueous solutions of CoCl_2 (1.9034 g), AlCl_3 (0.9657 g) and $(\text{NH}_2)_2\text{CO}$ (1.6817 g) at the molar ratio of 2:1:7 were mixed under magnetic stirring at about 97°C for 2 days under N_2 atmosphere. CoAl-LDH precipitate was separated by centrifugation, washed many times with deionized (DI) water and then ethanol, and dried at 60°C to obtain magenta-colored powder. Then, the intercalation of CoAl-LDH with dodecylsulfate (DS^-) was conducted by the reaction of an aqueous solution of sodium dodecylsulfate and an aqueous suspension of CoAl-LDH under magnetic stirring for ten minutes, and then transferred into Teflon line autoclave at hydrothermally heating of 120°C for 1 day (Ogawa and Hiramane, 2014). The loading amount of DS^- was adjusted to be 5 (0.8651 g), 10 (1.7303 g), 20 (3.4605 g) and 30 (5.1908 g) times of the theoretical AEC (anion exchange capacity ~ 300 meq/100 g, Wang and O'Hare, 2012) of the LDH. The modified CoAl-LDH was isolated by centrifugation, washed several times with DI water and then ethanol, and dried at 60°C to obtain coral-colored solid. The as-prepared hybrids were abbreviated as $\text{DS}_x/\text{CoAl-LDHs}$ where subscript x is referred to the ratio of the adding amount of DS^- to the AEC.

Removal of dye using the adsorbent

To evaluate the property of the organo $\text{DS}_x/\text{CoAl-LDHs}$ in comparison with pristine CoAl-LDH for eliminating toxic dye, the removal of rhodamine 6G, orange II and alizarin (Scheme 1) was examined at room temperature in the dark area. Each reaction, 50 mL dye solution (1:1 v/v of DI water and ethanol) with 20 ppm concentration and 10 mg of the adsorbent powder were mixed in beaker under magnetic stirring for 3 h in the dark. The adsorption reaction was conducted

under solution pH of rhodamine 6G, orange II and alizarin about 6.5, 5.0 and 6.3, respectively. During interval time of 5, 10, 20, 30, 45, 60, 90, 120, 150 and 180

min, the residual solution of dye was collected to examine the removal efficiency as the equation 1 by using UV-visible spectrophotometer.

$$\text{Removal efficiency (\%)} = \frac{C_0 - C_t}{C_0} \times 100 \quad 1$$

where C_0 (mg/L) is the initial concentration of dye and C_t (mg/L) is the concentration of dye at time t (min)

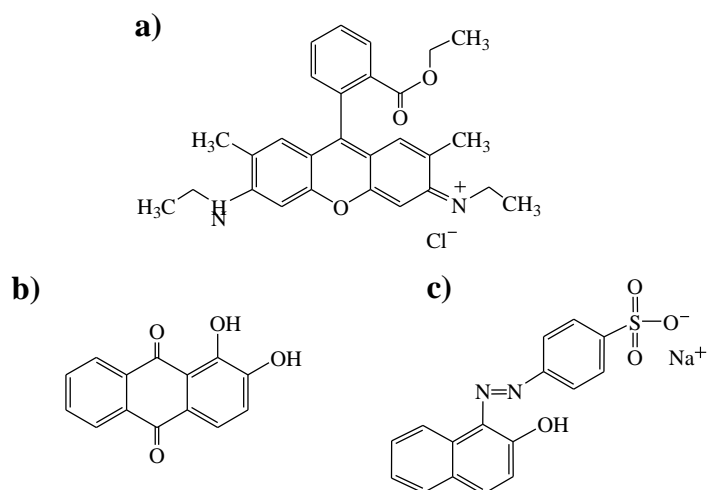
Characterization

Powder X-ray diffraction (XRD) patterns were collected on a Bruker D8 ADVANCE diffractometer using monochromatic $\text{CuK}\alpha$ radiation with $\lambda = 1.5418 \text{ \AA}$ operated at 40 kV and 40 mA. The data were collected at the scanning rate of 0.5 degree per minute over the region of 2-50 degrees. Infrared spectra of the samples were recorded on a Perkin Elmer Spectrum One FT-IR spectrophotometer by KBr disk method. UV-visible absorption spectra of the dye solutions were conducted in the wavelength range of 200-800 nm using a Shimadzu UV-1800 Pharmaspec UV-VIS spectrophotometer.

RESULTS AND DISCUSSION

To verify the intercalation of DS^- , the XRD patterns of $\text{DS}_x/\text{CoAl-LDH}$ s and pristine CoAl-LDH are displayed in Figure 1. The characteristic reflection of (003) correlating the size of carbonate (CO_3^{2-}) was observed at 2θ about 11.7° for all

samples with the basal spacing (d_{003}) of *ca.* 0.8 nm that calculated from Bragg's equation ($d = n\lambda/2\sin\theta$, Kong *et al.*, 2018), interpreting to the presence of carbonate in all samples (Scheme 2a). The basal spacings are summarized in Table 1. Commonly, carbonate generated in the reaction through CO_2 dissolved in water, possesses high affinity on positive surface of brucite-like structure due to small size and bidentate nature, resulting in the uncompleted substitution by DS^- . After adding the DS^- , the two basal spacings (d_{003}) of all products were obtained that calculated from Bragg's equation. The first d_{003} value was 0.76 nm, and the second one was 2.61 nm for $\text{DS}_5/\text{CoAl-LDH}$ and $\text{DS}_{10}/\text{CoAl-LDH}$, 3.12 nm for $\text{DS}_{20}/\text{CoAl-LDH}$ and 2.53 nm for $\text{DS}_{30}/\text{CoAl-LDH}$, which corresponded to the size of CO_3^{2-} and DS^- in the interlayer space respectively, indicating the segregation of DS^- and CO_3^{2-} in the interparticle level (Ogawa and Hiramane, 2014).



Scheme 1 Dye structures of (a) rhodamine 6G, (b) alizarin and (c) orange II

As seen in Figure 1, the d_{003} increased from 2.61 nm (Figure 1c-d) to 3.12 nm (Figure 1b) with low XRD peak intensity as increasing the loading amount of DS^- (x from 5 and 10 to 20), while x = 30 (Figure 1a) the d_{003} dropped to 2.53 nm with high XRD peak intensity. This result indicated the difference in nature of species (DS^- and CO_3^{2-}) in the interlayer space such as amount and arrangement. It was thought that the adding amount of DS^- (x = 30) enabled exchanging the intercalated carbonate more resulting in higher crystallinity as a result of high XRD peak ($d_{003} = 2.53$ nm) and low XRD peak ($d_{003} = 0.76$ nm) intensity. Taking the thickness of the LDH nanosheet (0.48 nm, Liu *et al.*, 2006) and the second d_{003} value into account, the expansion of the interlayer space of the products was determined to be 2.13, 2.13, 2.64 and 2.05 nm for $\text{DS}_5/\text{CoAl-LDH}$, $\text{DS}_{10}/\text{CoAl-LDH}$, $\text{DS}_{20}/\text{CoAl-LDH}$ and $\text{DS}_{30}/\text{CoAl-LDH}$, respectively (Table 1). This confirmed the

intercalation of DS^- in the interlayer space of the LDH with paraffin monolayer arrangement of DS^- in the interlayer space (Demel *et al.*, 2014; Okada *et al.*, 2014) as shown in Scheme 2b (Kong *et al.*, 2018), meanwhile some carbonate ions were also residual corresponding to the FTIR result (discussed later). The slight difference in the expansion of the interlayer space might be due to the variations in the amount of the intercalated water, carbonate and/or DS^- , as well as the flexibility of the DS^- long-chains. This study showed that the optimal amount of DS^- was 30 times of AEC, meanwhile Ogawa and Hiramane (2014) used DS^- up to 50 times of AEC for achieving intercalation into CoAl-LDH prepared by hydrothermal method (80 °C for 2 days). The difference in the adding amount of DS^- might be dependent on the presenting amount of the intercalated carbonate that varied by different preparation.

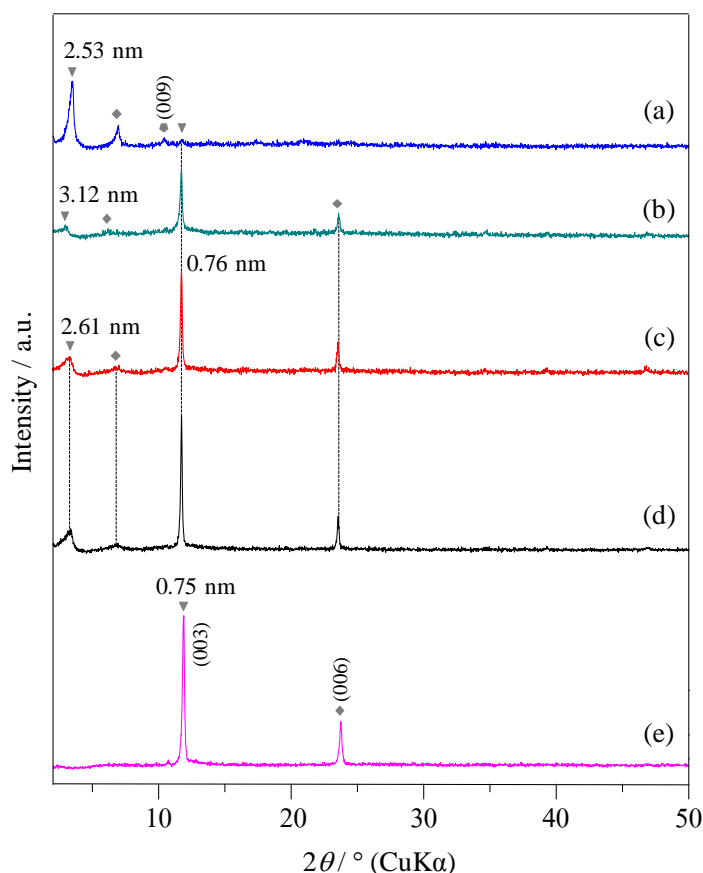


Figure 1 XRD patterns of (a) DS₃₀/CoAl-LDH, (b) DS₂₀/CoAl-LDH, (c) DS₁₀/CoAl-LDH, (d) DS₅/CoAl-LDH and (e) CoAl-LDH

Table 1 Basal spacing and interlayer expansion of all samples

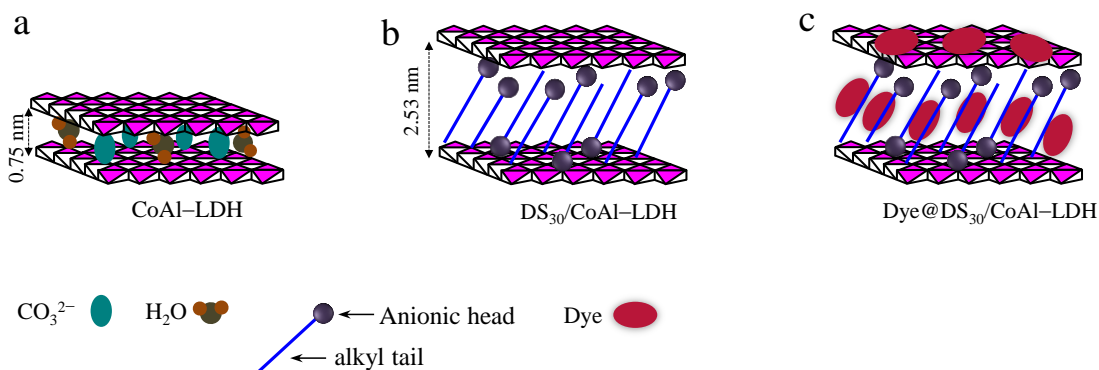
Product	d_{003} due to carbonate (nm)	d_{003} due to DS ⁻ (nm)	Interlayer expansion due to DS ⁻ (nm)
DS ₃₀ /CoAl-LDH	0.76	2.53	2.05
DS ₂₀ /CoAl-LDH	0.76	3.12	2.64
DS ₁₀ /CoAl-LDH	0.76	2.61	2.13
DS ₅ /CoAl-LDH	0.76	2.61	2.13
pristine CoAl-LDH	0.75	–	–

To verify the intercalation of DS⁻ in the interlayer space of CoAl-LDH and the substitution of the intercalated carbonate, the FTIR spectra of DS_x/CoAl-LDH together with CoAl-LDH and SDS were studied (Figure 2). The characteristic absorption bands of CoAl-LDH (Figure 2f) were corresponding to the hydrotalcite-like structure of the LDH counterbalanced with carbonate showing the vibration band about 1359 cm⁻¹ (Liu *et al.*, 2006). Also these characteristic bands were observed for all products (Figure 2b-e), interpreting

to the remaining host structure after intercalation reaction, which corresponded to the XRD result. Besides, the asymmetric and symmetric C–H stretching vibrations were also seen at 2925 and 2854 cm⁻¹ respectively, which shifted to higher energy region in comparison with those of SDS (at 2918 and 2849 cm⁻¹, Figure 2a), suggesting to the intercalation of DS⁻ in the hybrids by substitution of carbonate that evidenced by gradual disappearance of the band observed about 1359 cm⁻¹ (Figure 2). In the experiment, it was found

that increasing the adding amount of DS^- over 30 times of the AEC the small band due to residual carbonate was still appeared, supporting to the $d_{003} = 0.76$ nm,

as a result, the optimal amount of DS^- was $x = 30$ to eliminate carbonate from the interlayer space.



Scheme 2 Proposed arrangement of (a) CoAl-LDH, (b) organo $\text{DS}_{30}/\text{CoAl-LDH}$ in the absence of the intercalated carbonate and (c) dye adsorbed in the adsorbent

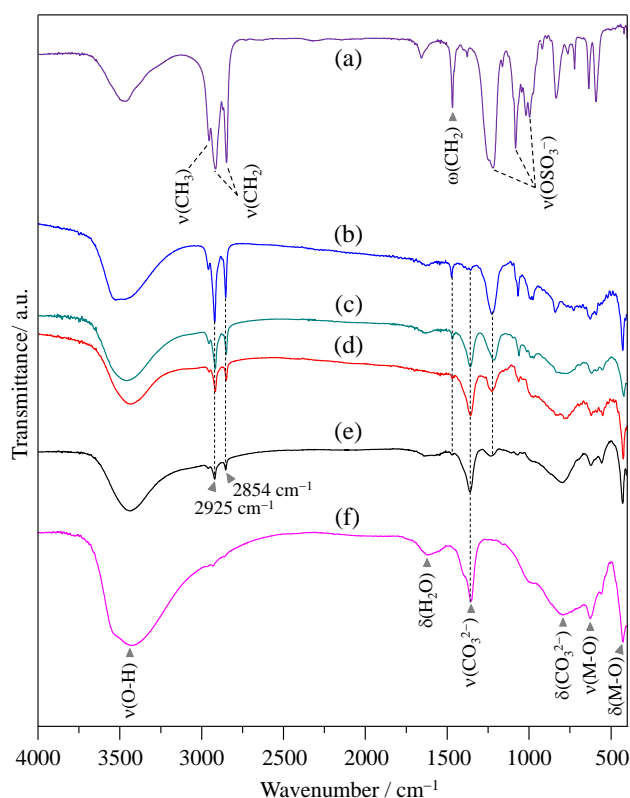


Figure 2 FTIR spectra of (a) sodium dodecylsulfate (SDS), (b) $\text{DS}_{30}/\text{CoAl-LDH}$, (c) $\text{DS}_{20}/\text{CoAl-LDH}$, (d) $\text{DS}_{10}/\text{CoAl-LDH}$, (e) $\text{DS}_5/\text{CoAl-LDH}$ and (f) CoAl-LDH

To solve the contamination due to dye in wastewater, the inorganic adsorbents, CoAl-LDH modified with dodecylsulfate ($\text{DS}_x/\text{CoAl-LDHs}$), were used to remove the toxic dyes in solution.

Among of the pollutants, rhodamine 6G, orange II and alizarin were used as the representative due to highlight of colorful pigment responsive for industrial market, and hardly-decomposable structure via bio

and chemical strategies. The maximum content of the dye adsorbed by the adsorbents was conducted for 60 min under magnetic stirring due to the equilibrium (Figure 3). As seen in Figure 3 and Figure 4, $DS_x/CoAl-LDHs$, where $x =$

5, 10, 20 and 30, showed the efficiency for removal of three dye species higher than $CoAl-LDH$ using as adsorbent, indicating to the effect of DS^- in the interlayer space.

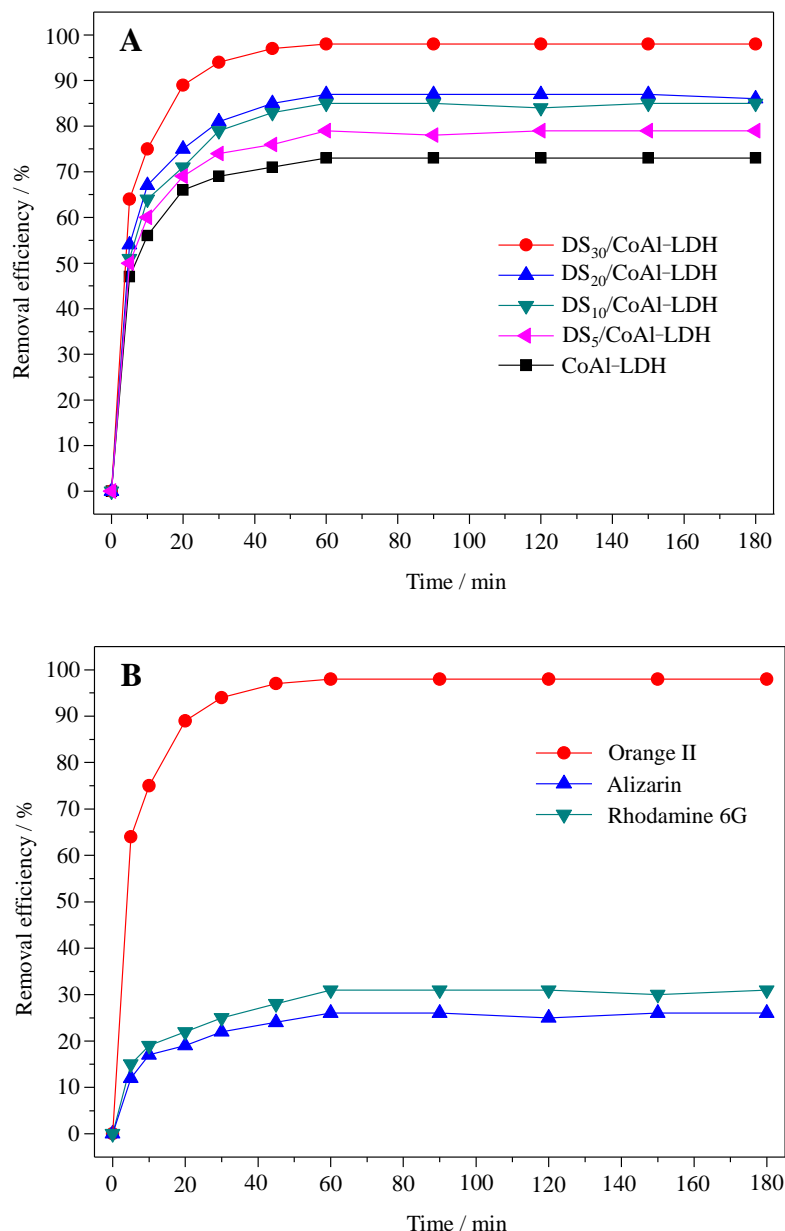


Figure 3 Effect of contact time for the adsorption of orange II using the inorganic adsorbents (A) and for the adsorption of orange II, alizarin and rhodamine 6G using $DS_{30}/CoAl-LDH$ (B)

In addition, the increase of the adding amount of DS^- in the adsorbent promoted the progress of the dye adsorption (Figure 3). This result was

because of the hydrophobic and hydrogen bonding interactions between the intercalated DS^- and the organic-dye structures (Kong *et al.*, 2018). Among of

three dyes possessing positive and negative charges, and neutral, the amounts of negatively charged orange II were mostly removed by 98%, and rhodamine 6G and

alizarin were adsorbed just 26% and 31%, respectively by using DS₃₀/CoAl-LDH as the adsorbent (Figure 3 and Figure 4).

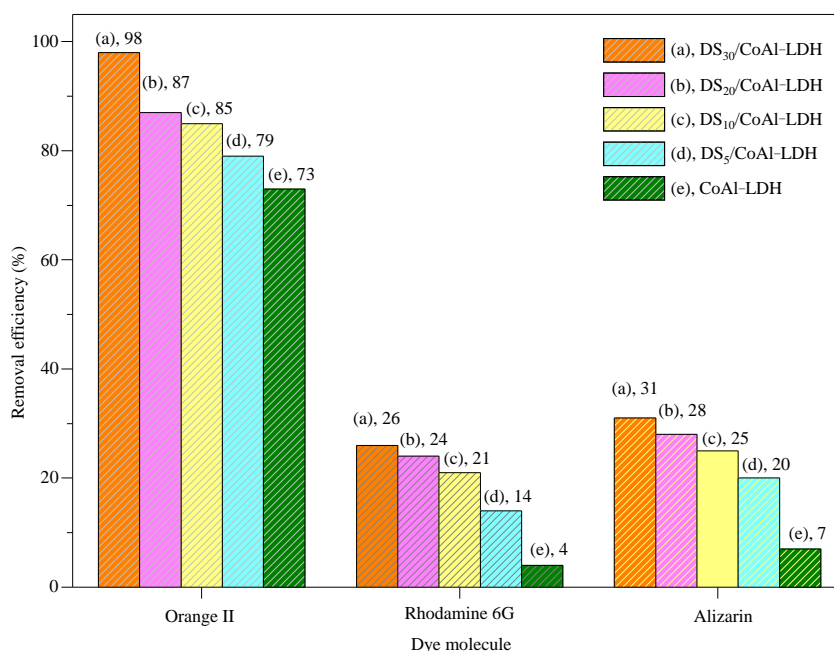


Figure 4 Removal efficiency of various dye molecules by using DS_x/CoAl-LDHs as adsorbent

This resulted mostly from the attractive force between negative-charged orange II and positive surface of the adsorbent, and larger adsorption of alizarin than that of rhodamine 6G might possibly due to the repulsion of positive-charged rhodamine 6G to the positive surface (Kong *et al.*, 2018) and the steric effect due to bigger size (Gamoudi and Srasra, 2019) to intercalate in the interlayer space as shown in Scheme 2c. Besides, the result showed that the electrostatic attraction was more essential role than that of the hydrophobic and hydrogen bonding interactions. We thought that the usage of DS₃₀/CoAl-LDH as the adsorbent for removing other anionic dye molecules should be further investigated.

CONCLUSIONS

The dodecylsulfate was intercalated into the interlayer space of CoAl-layered double hydroxide by hydrothermal conditions at 120 °C for 1 day. The optimal amount of

dodecylsulfate was 30 times of AEC of layered double hydroxide to eliminate carbonate in the interlayer space. The results from XRD patterns and FTIR spectra of DS₃₀/CoAl-layered double hydroxide confirmed the presence of most dodecylsulfate and remaining carbonate in the interlayer space due to the segregation. DS₃₀/CoAl-layered double hydroxide showed the best adsorbent and the potential for removing orange II from solution in comparison with alizarin and rhodamine 6G due to the major effect of the electrostatic interaction.

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