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Treatment of dye wastewater by *in situ* hybridization with Mg–Al layered double hydroxides and reuse of dye sludge

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ABSTRACT

A multi-anion dye (e.g. direct blending scarlet D-GLN) can intercalate into Mg–Al-layered double hydroxides (LDHs) which is confirmed from XRD, SEM, FTIR, DTG and ζ-potential analyses. The DGLN–LDHs hybrid with the hybridization amount 524 mg/g DGLN exhibits a high adsorption capacity to cationic dyes, e.g. 1064 mg/g for victorial blue B. The hybrid was used as a sorbent to treat cationic dye wastewater and the decoloration rate over 80% by adding only 5 g/dm^3. In addition, the resulting sludge was reutilized as a colorant filled in polymer materials and it exhibited the resistance to bleeding and fire. This work provides a new approach for the treatment of dye wastewater and reutilization of dye sludge.

1. Introduction

Synthetic dyes are widely utilized in textile, paper-made, dyeing, and other industries, leading to a large-scale development of dye industries. Consequently, a large amount of dye wastewater was produced. Without doubt, direct discharge of dye effluents will cause serious environmental problems, e.g. aesthetic color pollution, light penetration interference and toxicity to aquatic organisms [1–5]. Highly salty dye-wastewater is not suitable for microbiological process [4]. Flocculation is a simple and effective method but the resulting sludge disposal will bring out the secondary pollution. Advanced oxidation process is effective to decompose dye but unfavorable for carbon emission [6]. Eco-friendly treatment of dye wastewater is a challenging research to environmental scientists.

Layered double hydroxides (LDHs) is one of the superb anionic exchange materials [6], cheap and facile to prepare, and compatible to environment with a widespread application in agriculture, pharmaceuticals, detergent manufacturing and as food additives [7]. Recently, LDHs began to be used in the removal of anionic dyes [6,8–11]. However, less attention is paid on the further utilization of the LDHs–dye sludge. We found that the *in situ* LDHs–anionic dye sludge adsorbed strongly the organic compounds positively charged, e.g. basic or cationic dyes. A anionic dye–DGLN (Scheme 1) with five sulfonic groups was hybridized into the LDHs and the hybrid formed used as a sorbent to adsorb cationic dyes. In addition, the LDHs–anionic dye–cationic dye sludge was reused as colorant filled in polymer by referring to the previous work [12]. According to this work's cartoon illustrating in Fig. 1, the organic carbon in dye wastewater is immobilized and then reused so that the carbon emission reduction realized.

2. Experimental

2.1. Apparatus and materials

A photodiode array spectrometer (Model S4100, Scinco, Korea) with the Labpro plus software (Firmware Version 060105) was used to determine concentration of various dye solutions. Scanning electronic microscopy (SEM) (Model Quanta 200 FEG, FEI Co., USA) was used to measure the size and shape of materials. The FT-IR spectra of the hybrids were obtained, using an infrared spectrometer system (Model Equinoxss/hyperion 2000, BRUKER Co., Germany). The small-angle X-ray diffraction (SAXRD) (D/Max-2550 PC, Japan) was recorded using CuK radiation in the range of $\sim$1–10° 2θ for at a voltage of 30 kV and current of 50 mA and the

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Abbreviations: DGLN, direct blending scarlet DGLN; EV, ethyl violet; RBRK, reactive brilliant red K-2BP; BBBO, basic brilliant blue BO; LDHs–DGLN, a hybrid of LDHs and DGLN; Sample 5#, an anionic dye wastewater; LDHs–DGLN–3#, a suspending substance liquid prepared by sample 3# after treated by LDHs–DGLN; LDHs–DGLN–4#, a suspending substance liquid prepared by sample 4# after treated by LDHs–DGLN; AP, acrylic polymer coating; WAGGS, weak acidic green GS; MB, methylene blue; CR, congo red; VBB, Victoria blue B; Sample 1–4#, four cationic dye wastewater; LDHs–5#, a hybrid of LDHs and sample 5#; AP–LDHs–DGLN–3#, acrylic polymer coating filled LDHs–DGLN–3#.

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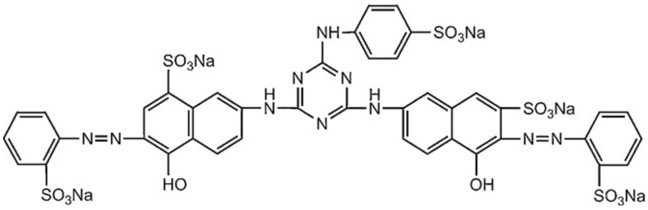


Scheme 1. Chemical structure of DGLN.

wide-angle X-ray diffraction (WAXRD) (Bruker D8 Advance, Germany) in range of ∼10–90° 2θ at 40 kV and 40 mA. Thermal analysis system (Q600 SDT Simultaneous DSC-TGA, TA instruments, USA) was used to determine TG of the materials. A ζ-potential instrument (Zetasizer Nano Z, Malvern, United Kingdom) was used to determine the surface potential of suspended substances. A inductively coupled plasma optical emission spectrometer (ICP-OES) (Optima 2100 DV, PerkinElmer, USA) was used to determine metal contents in materials. An automatic color measurement instrument (WSC-Y, Beijing, China) was used to determine the color parameters of paints filled with dye waste sludge. A spreader (QTG-A, Tianjin, China) was used to prepare the coating film (100–250 μm thickness) on paper.

The following dyes were dissolved in distilled water and their adsorptions investigated on with the LDHs–DGLN hybrid: RBRK (C.I. 18208, anionic dye), WAGGS (C.I. 61580, anionic dye), CR (C.I. 22120, anionic dye), EV (C.I. 42600, cationic dye), BBBO (C.I. 42595, cationic dye), MB (C.I. 52015, cationic dye) and VBB (C.I. 44045, cationic dye). Four cationic dye wastewaters (unknown wastewater) were sampled from Xingwu Dye Plant (Nantong, China) (sample 1#), Zhejiang Runtu Corp. (Shangyu, China) (sample 2#), Jinjiang Chemical Plant (Hangzhou, China) (sample 3#) and Xinghua Chemical Corp. (Yancheng, China) (sample 4#). An anionic dye wastewater (sample 5#) was sampled from Shuanghong Chemical Corp. (Jinhua, China). The epoxy topcoating wall A and B were purchased from Shanghai Lvjia Waterborne Coatings Corp. A weathershield Plus paint (AP) was purchased from Dulux in Shanghai.

2.2. Synthesis and characterization of the LDHs–DGLN hybrid

The LDHs–DGLN hybrid was prepared by imitating the coprecipitation method [13]. A mixture containing $Al(NO_3)_3{\cdot}9H_2O$ (15.0 g), NaOH (12.8 g) and DGLN (18.1 g) was dissolved in 2500 mL of deionized water (Solution A). 400 mL of Solution B containing 20.5 g $Mg(NO_3)_2{\cdot}6H_2O$ was added dropwise into Solution A under vigorous stirring for 2 h. The mixture was adjusted to pH 10 with a HNO_3 solution. After settling for 10 h, the DGLN concentration in the supernatant was determined at 540 nm by spectrophotometry. The suspension was centrifugated, washed with deionized water. Then it was dried and ground into power. The SEM, FTIR, and TGA of the powder were measured. The Al and Mg contents were determined by ICP-OES after dissolving the powder in a HCl solution.

2.3. Adsorption of dyes

The LDHs or the LDHs–DGLN hybrid (0.1%) was added into cationic dyes (e.g. EV, BBBO and MB) and anionic dyes (e.g. RBRK, WAGGS and CR) solutions, respectively. The phenomenon of the selective adsorption experiment was observed. Then, 0.05% of the LDHs–DGLN hybrid was added into the WAGGS solutions from 40 to 280 μM and EV solutions from 200 to 540 μM. Into the VBB solutions series from 100 to 290 μM, 0.01% of the LDHs–DGLN hybrid was added. After settlement, all the mixtures were centrifugated and the dye concentrations in the supernatants determined by spectrophotometry. The experimental data of WAGGS, EV and VBB adsorbing onto the LDH–DGLN hybrid were fitted by Langmuir isotherm [8,14].

$$\frac{1}{q_e} = \frac{1}{K_l q_\infty c_e} + \frac{1}{q_\infty}$$

where q_e (mg/g) was the adsorption amount of dye, q_∞ (mg/g) the saturation adsorption amount of dye, c_e (mmol/L) the equilibrium concentration of dye, and K_l the adsorption constant.

EV was chosen for investigating the effects of the experimental conditions, e.g. pH, temperature, ionic strength and equilibration time. HCl and NaOH solutions were used to adjust pH of the liquids. The temperature test, e.g. 25, 30, 35, 40, 45, and 55 °C, was carried out in a constant temperature water bath for 15 min. 1 M NaCl was used to adjust ionic strength of the liquids from 0 to 0.2 M. The sorption equilibration time was carried out by sampling at 0, 2, 5, 8, 11, 15, 20, 25, 30 and 45 min. The liquids were centrifugated and the dye concentration in the supernatants determined.

2.4. In situ treatment of direct dye wastewater

According to Section 2.2, an anionic dye wastewater (sample 5#) was used as the reactant instead of DGLN. Both $Al(NO_3)_3{\cdot}9H_2O$ (1.5 g) and NaOH (1.3 g) were mixed in 100 mL of the wastewater (solution A). Solution B containing $Mg(NO_3)_2{\cdot}6H_2O$ (2.1 g) was prepared with 100 mL of deionized water. The preparation was the same as that of Section 2.2. The suspending substance liquid (LDHs–5#) was used to treat cationic dye wastewater.

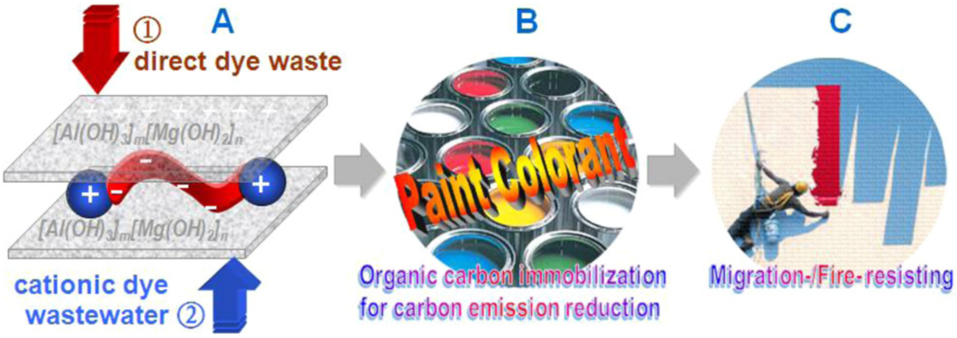


Fig. 1. Cartoon illustration of formation and reuse of dye sludge. (A) Direct dye waste intercalating into LDHs and then adsorbing cationic dye. The blue globe between layers represents cationic dye, and the red ribbon represents direct dye (e.g. DGLN). (B) Dye sludge as colorant mixed into paint. (C) Use of paint filling with dye sludge. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

2.5. Reuse of dye sludge

2.5.1. Used as sorbent

The LDHs–DGLN hybrid was used to treat cationic dye wastewater by adding into samples 1#–4#. The suspension from samples 3# and 4# were named as LDHs–DGLN–3# and LDHs–DGLN–4#. In addition, sample 2# was treated by adding LDHs–5# from 0.04% to 0.8%. Thus, the adsorption capacities of these hybrids formed from DGLN and dye wastewater were compared. The colority of the supernatants was measured by spectrophotometry at 651 nm for sample 1#, 510 nm for sample 2#, 440 nm for sample 3# and 544 nm for sample 4# [15]. The decoloration rate was calculated by the relation:

$$\mathit{Decoloration\ rate}\,(\%) = \frac{A_0 - A}{A_0} \times 100$$

where A_0 and A were absorbances of the supernatant before and after treating.

2.5.2. Used as paint colorant

5 mL of epoxy topcoat wall A was mixed into 25 mL of epoxy topcoat B, and 2 mL of LDHs–DGLN–3# (1.61%) or LDHs–DGNL–4# (0.51%) added under stirring for 45 min. These mixtures were painted on the glass slides and then dried for 16 h at room temperature. The color of the glass slide was measured with an automatic color measurement instrument (WSC-Y, Beijing, China).

2.5.3. Migration resistance test

The glass slides prepared according to Section 2.5.2 were immersed into 2% HCl and 2% NaOH for over 24 h. After soaking, every glass slide was washed with deionized water and dried, and the color of the glass slides was measured.

2.5.4. Fire resistance test

Fire resistance test was referred to the standard of burning behavior of flame-retardant paper [16]. Both LDHs-only and LDHs–DGLN–3# were added into AP according to the mass ratio at 0.36:0.02:1. A blank sample was prepared by mixing only the weathershield plus paint. The paint prepared was spread evenly on a sheet of paper with a spreader with 250 μm of thickness. After dried for 5 h at 65 °C, several rectangle paper scrips were cut out and hung over a alcohol burner. They were ignited for 2 s from the paper bottom. The burning time and status of the paper scrips were observed and recorded.

3. Results and discussion

3.1. Formation and characterization of LDHs–DGLN hybrid

In the LDHs–DGLN hybrid, DGLN binding to LDHs was found to be 524 mg/g when the initial mole ratio of Mg^{2+} to Al^{3+} and DGLN was 8:4:1.5 at pH 10 (refer to Fig. S1). The mass ratio of Mg to Al and DGLN in the LDHs–DGLN hybrid was determined to be 8:3.5:1.2, which approached to the initial mole ratio. It indicates a high reaction efficiency of the reactants. The hybrid began to dissolve when pH was less than 4 (refer to Fig. S2). Elemental analysis showed that the LDHs is the Mg–Al–NO_3 LDHs.

From XRD (Fig. 2B-1), the basal d_{003} space (8.4792 Å) of LDHs is similar to that reported [8], and as 2 times high as d_{006} and 3 times high as d_{009}, which is characteristic to the hydrotalcite-like phase [17]. The broaden and weaken of the (0 0 3) peak of curve 2 (Fig. 2B) indicates that the hybridization of DGLN makes the particles less order. Curve 2 exhibits the same peak locations as curve 1. It means that the main structure of the hybrid remains unchange after the hybridization of DGLN. In addition, a new broad and weak peak is found at 2θ of ca. 4.85° with d space of ca. 18.2 Å (Fig. 2A-2). It is similar to the intercalation of acid blue 9 into the Mg–Al–CO_3 LDHs [11]. The d space at $2\theta = 4.85^\circ$ could correspond to the basal space of a new less-ordered layered material formed during the intercalation of DGLN into LDHs precursor. Considering 4.8 Å of thickness for the brucite layer [9], the basal space of 18.2 Å corresponds to an interlayer space of 13.4 Å. According to the three-dimensional size of DGLN (estimated as 28 Å × 19 Å × 8 Å), it is probably intercalated in the oblique direction of y-axis (Fig. 3).

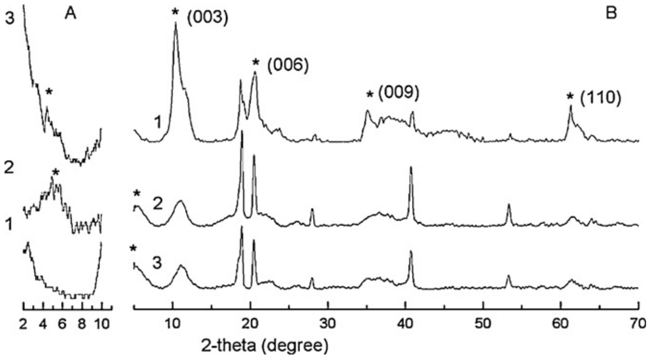
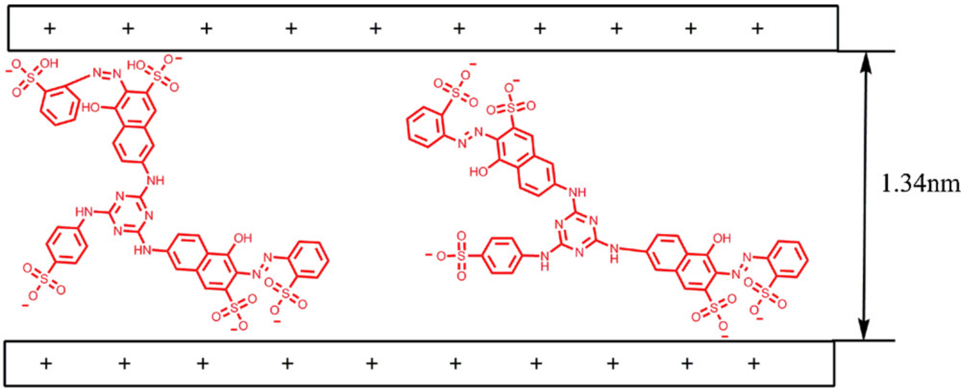


Fig. 2. XRD of LDHs and LDHs hybrids. (A) Small angle X-ray diffraction (SAXRD) of LDHs (curve 1), the LDHs–DGLN hybrid (curve 2) and the LDHs–DGLN–EV (curve 3). (B) Wide angle X-ray diffraction (WAXRD) of LDHs (curve 1), the LDHs–DGLN hybrid (curve 2) and the LDHs–DGLN–EV (curve 3).

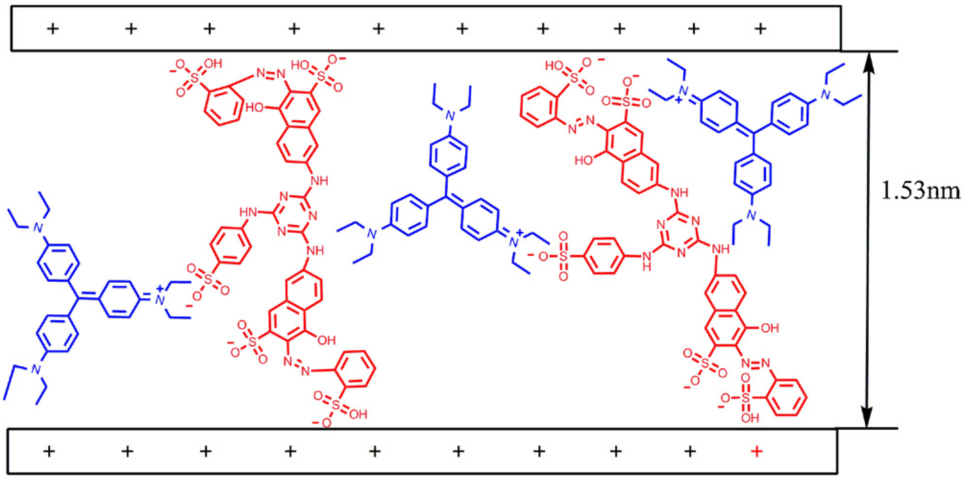
The intercalation of DGLN is confirmed by FT-IR spectroscopy. The LDHs exhibits a broad band at 3460 and 3550 cm^{-1} assigned to ν_{O-H} stretching vibration of the hydroxyl group, 1384 cm^{-1} for the characteristic stretching vibration of nitrates, and 448 cm^{-1} for O–Mg vibration [18,19] (Fig. 4). The characteristic bands of DGLN appear at 1602, 1474, 1383, 1237 and 1192 cm^{-1} due to the C=C vibration band of benzene ring (1602 and 1474 cm^{-1}), the C–H band of benzene ring (1383 cm^{-1}), ν_{C-O} bend vibrations of aryl ether (1237 cm^{-1}) and the stretching band of sulfur–oxygen groups (1192 cm^{-1}). In the FT-IR of the LDHs–DGLN hybrid, all the characteristic bands corresponding to DLGN and LDHs appear on the right positions. However, the vibrations of sulfonate group around 1192 cm^{-1} for the intercalate is shifted slightly to the lower frequency region at 1181 cm^{-1} compared to DGLN, which clearly indicated that the electrostatic interaction is present between DGLN and LDHs [9].

From DTG curve 1 (Fig. 5B), LDHs showed three major weight loss peaks around 146, 392 and 468 °C, corresponding to water evaporation, release of interlayer anions, the dehydroxylation of the inorganic hydroxide and formation of Mg/Al oxides [20,21]. DGLN has three major weight loss peaks around 76, 307 and 430 °C assigned to water evaporation, thermal decomposition of DGLN and release of carbon dioxide [10]. However, the LDHs–DGLN hybrid presents three different weight loss peaks. The first weight loss around 109 °C in curve 3 is between 76 and 146 °C, suggesting that the intercalation of DGLN makes the removal of interlayer water easy. The weight loss peaks was postponed to 444 and 618 °C in comparison with those of DGLN around 307 and 430 °C, which indicates that the thermal stability of DGLN is improved after intercalating into the LDHs. In addition, the dehydroxylation of the inorganic hydroxide layers delayed the weight loss and showed a less weight loss between 200 and 650 °C by comparing with DGLN and LDHs (Fig. 5A). Therefore, the electrostatic interaction between DGLN anion and hydroxide layer is present.

The ζ-potential measurement indicated that the intercalation of DGLN into LDHs changed the surface electric potential from +2.87 of LDHs to −19.7 mV of the hybrid. It indicated that the LDHs–DGLN hybrid will adsorb cationic organic compounds.



A: Intercalation of DGLN into LDHs



B: Sorption of EV by LDHs-DGLN

Fig. 3. Intercalation of dyes into LDHs. (A) Intercalation of DGLN into LDHs. (B) Sorption of EV by LDHs–DGLN.

3.2. *Adsorption of dyes with the LDHs–DGLN hybrid*

In order to reveal the adsorption selectivity of the LDHs–DGLN hybrid, anionic dyes, e.g. RBRK-2BP, WAGGS and CR and cationic dyes, e.g. EV, BBBO and MB were treated with the hybrid (refer to Fig. S3). The LDHs–DGLN hybrid exhibited much more obvious adsorptions to all these dyes, specially anionic dyes than LDHs-only. WAGGS, EV and VBB were selected for illustrating the adsorption mechanism and the experimental data fitted by the Langmuir

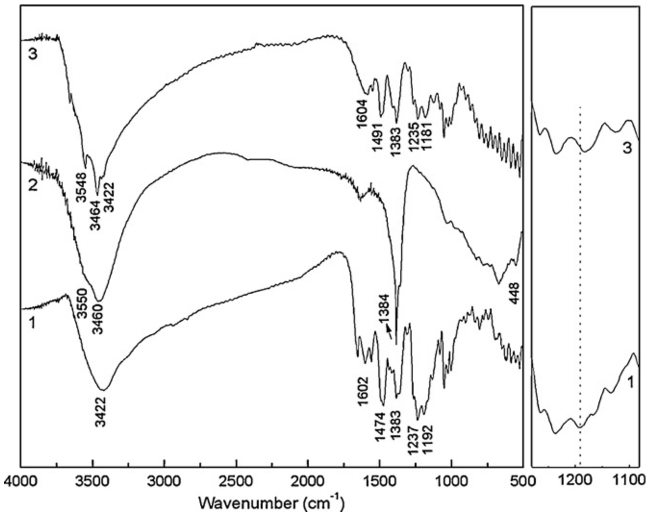


Fig. 4. FTIR of DGLN (1), LDHs (2) and LDHs–DGLN hybrid (3).

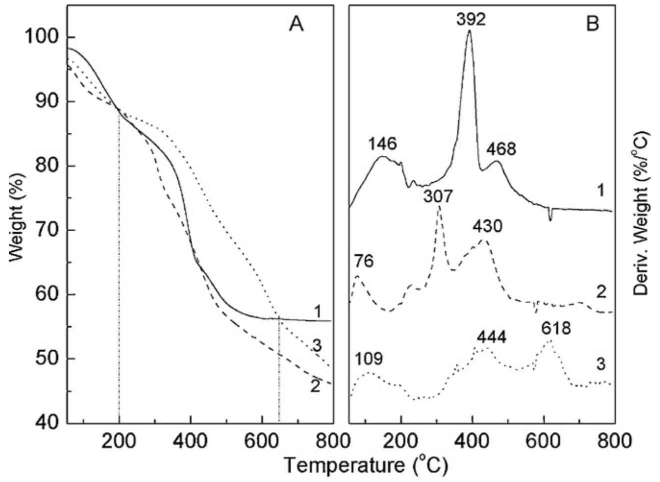


Fig. 5. TG (A) and DTG (B) of LDHs (curve 1), DGLN (2) and LDHs–DGLN hybrid (3).

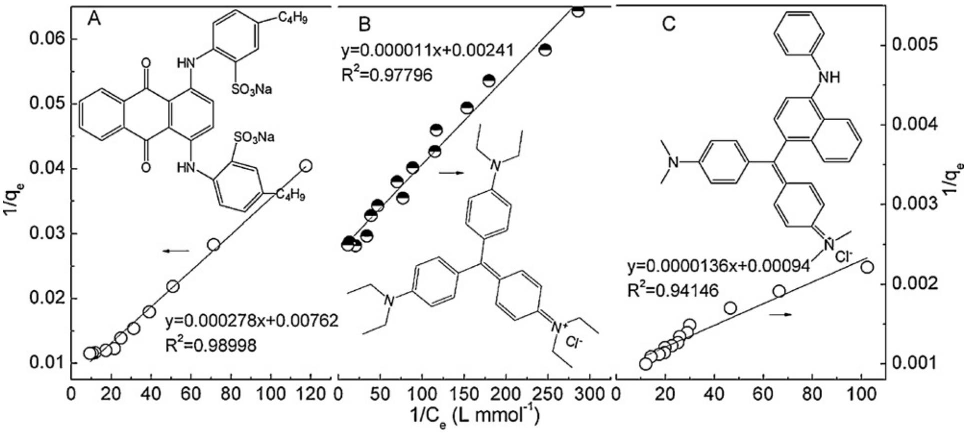


Fig. 6. Plots q_e^{-1} vs. c_e^{-1} of WAGGS (A), EV (B) and VBB (C) treated with the LDHs–DGLN hybrid as adsorbent.

isotherm (Fig. 6) (The impact of the initial concentration of the three dyes on q_e was seen in Fig. S4.) The q_e^{-1} of the three dyes are linear with their c_e^{-1} (Fig. 6). The q_∞ values of WAGGS, EV and VBB were calculated to be 131, 415 and 1064 mg/g (K values were 27.5, 219.1 and 69.1 L/mmol), i.e. their molar ratios to the DLGN intercalated in the hybrid are 0.4:1, 1.6:1 and 3.6:1. Apparently, both EV and VBB were adsorbed strongly in comparison with WAGGS. Therefore, the electrostatic adherence plays a major role. In addition, the sorption of WAGGS indicated that not all NO_3^- anions were replaced by DGLN during the LDHs–DGLN hybrid synthesis. It is possible due to steric effect of DGLN. Just the steric effect caused the sorption of EV being less than VBB. It can also be confirmed from the ζ-potentials from −19.7 mV of the LDHs–DGLN hybrid to −13.6 mV of the LDHs–DGLN–EV hybrid.

From XRD of the LDHs–DGLN–EV hybrid (Fig. 2B), curve 3 has the same peaks location as curve 1. A new reflection peak is also found at 2θ of ca. 4.40° (Fig. 2A), which gives a d space of ca. 20.1 Å. The sorption of EV increases the basal space ($d_{\text{LDHs–DGLN}}$ 18.2 Å), revealing that EV might intercalate into hydroxide layers. The SEM of the LDHs–DGLN–EV hybrid has not changed compared with that of the LDHs–DGLN (Fig. 7) and its FT-IR approves the intercalation of EV (refer to Fig. S5). The C=C vibration of benzene ring of EV around 1578 cm^{-1} is shifted slightly to the higher frequency region compared to that of EV. Thus, the electrostatic interaction is present between EV and the LDHs–DGLN hybrid (Fig. 3C).

Effects of pH, ionic strength, temperature and time on adsorption of EV were investigated (Fig. S6). The sorption amount of EV remained constant between pH 5 and 8 and increased slightly at pH 8–11 (refer to Fig. S6A, B). The sorption of EV increased slowly with increase of electrolyte (NaCl solution). Thus, it is favorable for the treatment of highly salty dye wastewater. With increase of temperature from 26 to 55 °C, the removal rate of EV increased slightly. A high temperature benefits to the sorption of EV on the hybrid (Fig. S6C). Up to 90% of EV adsorption occurred within the initial 10 min, and equilibrium appeared after 30 min (Fig. S6D).

The adsorption capacity of the LDHs–DGLN hybrid was compared with the other sorbents [15,22–24] (Table 1). The capacity of EV was as 4 times high as that of activated carbon cloth and the $CaCO_3$–APRB hybrid, and 2 times high as that of the pyrolyzed sewage sludge. The LDHs–DGLN hybrid can adsorb 1064 mg/g VBB, being much higher than fly ash does.

3.3. Treatment of cationic dye wastewater

Four kinds of cation dye wastewaters with colority 22500 (1#), 32485 (2#), 91234 (3#) and 1736 (4#) were treated with the LDHs–DGLN hybrid. The wastewater colority decreased with increase of the hybrid (Fig. 8). When 0.5% of the sorbent dosage was added, the decoloration rates of samples 1#, 2# and 4# were 86.5, 86.8 and 80%. When only 1% was added, the decoloration rate of sample 3# reached 80%. The initial colority of sample 3# is much

Table 1
Adsorption capacities of various sorbents to cationic dyes.

Adsorbent	Adsorbate	Capacity (mg/g)
LDHs–DGLN hybrid	EV	415
LDHs–DGLN hybrid	VBB	1064
Fly ash	VBB	0.18
$CaCO_3$–APRB hybrid	EV	112
Regenerated bleaching earth	EV	55
Pyrolyzed sewage sludge	EV	200
Activated carbon cloth	EV	100

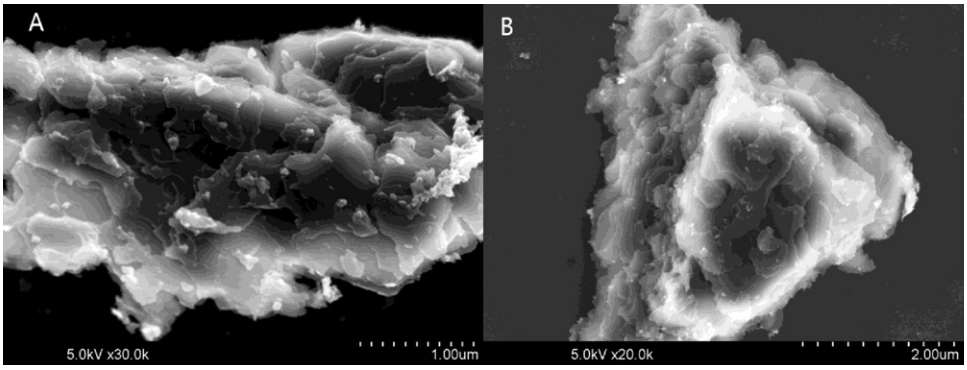


Fig. 7. SEM of LDHs hybrids. (A) SEM of the LDHs–DGLN hybrid. (B) SEM of the LDHs–DGLN–EV hybrid.

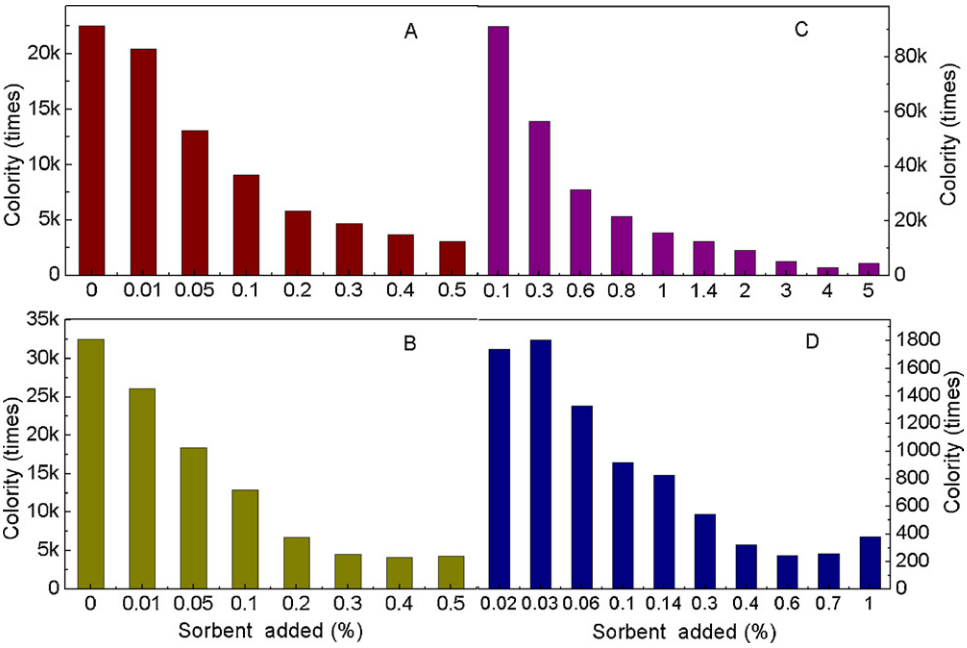


Fig. 8. Treatment of cationic dye wastewaters with the LDHs–DGLN hybrid. Wastewater samples 1# (A), 2# (B), 3# (C) and 4# (D).

more than the others but its decoloration rate is similar to that of the others. The LDHs–DGLN hybrid is efficient in treatment of cationic dye wastewater, especially for concentrated dye wastewater.

3.4. Treatment of anionic and cationic dye wastewaters

The DGLN binding to LDHs plays an important role in the sorption of cationic dyes. In order to reuse dye waste, an anionic dye wastewater (sample 5#) (colority, 10,000) was tried as the reactant instead of DGLN to prepare a LDHs–waste dye hybrid according to the synthesis Section 2.2. The *in situ* treatment of sample 5# exhibited a high removal rate 99.1% of colority. The resulting suspending substance (LDHs–5#) was used as sorbent to treat sample 2#. The decoloration rate of sample 2# reached 60% when only 0.6% of LDHs–5# was added (refer to Fig. S7). When LDHs–5# was added up to 1%, the removal rate of the color substances in sample 2# approached to 100%. In this way, anionic dye wastewater could be reused to prepare a sorbent of cationic dye wastewater by means of the *in situ* hybridization with LDHs.

3.5. Reuse of dye sludge as colorant

Dye sludge is often disposed by burning and landfilling. It may result in the secondary pollution to air and ground water. In fact, the dye sludge immobilizing with LDHs could be reutilized as colorant filled in polymer. Two factors of colorant, such as thermal stability and migration resistance were investigated. The thermal stability of DGLN improved was confirmed from the DGT analyses above. Migration of a colorant often occurs when it is dispersed into the polymer matrix [25]. To determine the migration, the epoxy paints filled with the LDHs–DGLN–3# and LDHs–DGLN–4# were immersed into acid (2% HCl) and alkaline (2% NaOH) media for 24 h. No extraction of colorant occurred from the epoxy paint. Thus, such two dye sludges exhibited a resistance to acid and alkaline.

In fact, LDHs is still used as a fire-resistance material added into plastic [26]. In order to investigate the fire-resistance effect of the LDHs–dye composite filled in AP, the burning test of AP paper scrips was carried out. The results indicated that the burning time was 25.1 ± 1.8 s for AP paper scrips and 29.0 ± 2.4 s for the AP paper scrips filling with LDHs–DGLN–3# (95% confidence level, 33 parallel samples per group). Thus, the burning time of AP paper scrips filled with the LDHs–DGLN–3# delayed for 4 s. In addition, 68% of the paper scrips with AP–LDHs–DGLN–3# extinguished themselves after ignition but only 3% of the paper scrips with AP-only appeared the same case. Therefore, the LDHs–dye sludge is feasible to reuse as color flame retardant.

4. Conclusions

The *in situ* hybridization of DGLN with LDHs was performed and the intercalation of DGLN reached 524 mg/g. The hybrid exhibits a highly effective adsorption capacity to cationic dyes, e.g. 415 mg/g EV and 1064 mg/g VBB. When it is used to treat dye wastewater, over 95% of color substances was removed. The *in situ* hybridization of anionic dye wastewater with LDHs is suggested to prepare the absorbent of cationic dyes. When the LDHs–dye sludge was reused as colorant, it exhibits a good resistance to fire and improves the thermal stability of the colorant. From dye wastewater treatment to reuse of dye sludge, a great deal of organic carbon was immobilized being favorable for carbon emission reduction.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.cej.2011.06.076.

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