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Dyes adsorption using a synthetic carboxymethyl cellulose-acrylic acid adsorbent

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ABSTRACT
Removal of noxious dyes is gaining public and technological attention. Herein grafting polymerization was employed to produce a novel adsorbent using acrylic acid and carboxymethyl cellulose for dye removal. Scanning electron microscopy and Fourier-transform infrared spectroscopy verified the adsorbent formed under optimized reaction conditions. The removal ratio of adsorbent to Methyl Orange, Disperse Blue 2BLN and malachite green chloride reached to 84.2%, 79.6% and 99.9%, respectively. The greater agreement between the calculated and experimental results suggested that pseudo second-order kinetic model better represents the kinetic adsorption data. Equilibrium adsorptions of dyes were better explained by the Temkin isotherm. The results implied that this new cellulose-based absorbent had the universality for removal of dyes through the chemical adsorption mechanism.

Introduction

Dyes are widely used in many industries such as textile, cosmetics, paper printing, leather and plastic industry (Badruddoza et al., 2010; Goel et al., 2011). Dyes are a relatively large group of organic chemicals classified based on their molecular structure as azobenzene, anthraquinone, or triphenylmethane dyes. Methyl Orange (MO; C.I. 35780), Disperse Blue 2BLN (DB; C.I.63285), and malachite green chloride (MG; C.I. 42000) are frequently used dyes representing the above classes. These dyes are difficult to degrade because of their complex aromatic structures and can cause allergy, dermatitis, irritation and even to cancer in humans (Bhattacharyya and Sharma, 2004; Xu et al., 2006). Residual and unspent dyes are usually discharged into the environment, thereby causing pollution problems (Deng et al., 2009). With the increasing concern on environment protection, removal of such dyes is gaining public and technological attention (Yan et al., 2011).

For dye removal from wastewater, methods such as sorption (Duran et al., 1999), flocculation (Gautam et al., 2011), oxidation (Mantzavinos and Psillakis, 2004), and electrolysis (Ruan et al., 2010) are common methods. Given that sorption can transform dyes from the effluent to a solid phase, this method is generally regarded as an effective technique for dealing with wastewater dye. Among conventional adsorbents, activated carbon has been widely investigated and used for dye adsorption from various effluents (Deng et al., 2009). Nevertheless, its application is limited because of its high cost and recalcitrant (O’Connell et al., 2008). In recent years, interest in finding an effective alternative to activated carbon such as starch (Wang et al., 2010), cellulose (Wojnárovits et al., 2010), chitosan (Wang et al., 2011) and lignin (Silva et al., 2011) is growing.

Cellulose is one of the most abundant natural substances in nature. However, the compact and inactive molecular
structure of cellulose requires it to be modified to improve its hydrophilicity as an adsorbent for dye removal. To date, the cellulose-based materials are prepared by carboxymethylation (Yan et al., 2011), grafting (Wang and Wang, 2008) and cross-linking (Liu et al., 2009a). Among the cellulose-based materials, carboxymethyl cellulose (CMC) is a representative water-soluble cellulose derivative. By itself or grafted with other water-soluble compounds, CMC is used as an adsorbent for dye sorption from various wastewaters systems (Yan et al., 2011; Bao et al., 2011). Several reports have also indicated that a particular kind of cellulose-based material can remove dyes with different molecular structures.

In this study, CMC prepared by our previous research (Zhang et al., 2011) was used as the basic adsorption material to prepare a novel adsorbent. This absorbent was produced by grafting acrylic acid (AA) along the chains of CMC to improve the mechanical properties, swelling capacity and dyes adsorption capacity of CMC. MO, DB and MG were chosen to investigate the adsorption behavior and universality of CMC-AA to the dyes. The preparation method of CMC-AA adsorbent has the advantage of lower temperature, short time and cheap raw material, and the CMC-AA adsorbent has high adsorption to multiple dyes, therefore, it can effectively reduce the production and application cost. The present research will provide the technical basis for preparation and application of CMC-based adsorbent.

1 Materials and methods

1.1 Materials

CMC used (with the degree of substitution of 0.77) was prepared according to our previous research (Zhang et al., 2011). AA, ammonium persulfate (APS) and N,N-methyl-enebisacrylamide (MBA) were supplied by Tianjin Reagent Corp (Tianjin, China). Other agents used were all of analytical grade and all solutions were prepared with distilled water.

1.2 Preparation of CMC-AA adsorbent

A series of samples were prepared according to the modified procedure (Liu et al., 2009b). A 2-g of CMC was dissolved with 50 mL distilled water in a 300 mL three-neck flask, equipped with a mechanical stirrer, a condenser and a nitrogen line. After purged with nitrogen for 15 min, 0.02 g of APS was added to flask for initiating radicals. Then 4–20 g of AA containing 0.006 g of MBA, neutralized to the certain pH with the solution of sodium hydroxide, was added. The grafting reaction was then performed in the water bath at 50–90°C for 2–6 hr. The gaining product was dried to a constant weight at 140°C and milled to particle size about in 0.40 mm. Grafting efficiency (GE), showing the formation of CMC-AA adsorbent but for homopolymer and in direct proportion to absorbency (Kuwabara and Kubota, 1996), was determined according to the reported method (Samaha et al., 2005). The gaining CMC-AA product was extracted for 4 hr to remove homopolymer by acetone via Soxhlet extractor. After the CMC-AA copolymer dried at 80°C, the GE was calculated as the following equation:

\[ GE = \frac{W_2}{W_1} \times 100\% \]

where, \( W_1 \) and \( W_2 \) are the weight of total product and the weight of CMC-AA only grafted on CMC (after acetone extraction), respectively.

1.3 Characterization of CMC-AA adsorbent

The FT-IR spectra of samples were taken in KBr pellets using an AVATAR360 FT-IR spectrophotometer (Nicolet, USA). The surface morphology of the adsorbent was examined using scanning electron microscopy (JSM-6490, JEOL, Japan). A sputter coater was used to pre-coat conductive gold onto the fracture surfaces before observing the microstructure at 30 kV. The surface analyses and porosity were determined from the adsorption-desorption isotherm of nitrogen at −196°C (Micromeritics ASAP2020). For area and pore calculations, the reported methods were used (Deng et al., 2009).

1.4 Adsorption studies of CMC-AA to MO, DB and MG

Adsorption studies were carried out in triplicate using the batch adsorption method. All of experiments were performed in the 100 mL of dyes solutions (MO and MG 30 mg/L, DB 200 mg/L) with 20 mg of CMC-AA adsorbent for 60 min at 25−50°C. The pH of the dye solutions were ranged from 2.0 to 10.0. After the equilibrium, the suspensions were centrifuged at 10,000 rpm for 5 min. The final concentration of dyes which remained in the batch adsorption method. All of experiments were performed in the 100 mL of dyes solutions (MO and MG 30 mg/L, DB 200 mg/L) with 20 mg of CMC-AA adsorbent for 60 min at 25−50°C. The pH of the dye solutions were ranged from 2.0 to 10.0. After the equilibrium, the suspensions were centrifuged at 10,000 rpm for 5 min. The final concentration of dyes which remained in the solution was determined by visible spectrophotometer. Absorbance measurements were made at the maximum wavelength of 464, 567 and 617 nm for MO, DB and MG respectively. The dyes removal (R) by the CMC-AA adsorbent was calculated by Eq. (2).

\[ R = \frac{C_0 - C_t}{C_0} \times 100\% \]

where, \( C_0 \) and \( C_t \) are the initial and residual concentration of dyes in the suspension.

1.5 Kinetic and equilibrium models

Adsorption kinetic studies can provide valuable information on the mechanism of the adsorption process. So the three kinetic models were used to evaluate the adsorption kinetic of dyes.

Pseudo first-order:

\[ q_t = q_e(1 - \exp(-k_1t)) \]
where, \( q_e \) (mg/g) and \( q_i \) (mg/g) are the amounts of adsorption dye at equilibrium and at time \( t \) (hr), \( k_1 \) (hr\(^{-1}\)) is the pseudo first-order rate constant.

Pseudo second-order:

\[
q_t = \frac{k_2q_e^2t}{1 + k_2q_e^2t}
\]

(4)

where, \( k_2 \) (g/(mg-hr)) is the pseudo second-order rate constant.

Intra particle diffusion:

\[
q_t = k_i(t^{1/2}) + C
\]

(5)

where, \( k_i \) (mg/(g-sec)) is intraparticle diffusion rate constant and \( C \) is constant. \( C \) values give information about the thickness of boundary layer and is increasing with increase of initial concentration.

The thermodynamic assumptions of adsorption isotherms and their estimated parameters provide the insight for both the properties of the surface and the mechanism of adsorption. So the three isotherm models were used to evaluate the equilibrium of adsorption.

Langmuir isotherm model:

\[
q_e = \frac{Q_{\max}KLC_e}{1 + KLC_e}
\]

(6)

where, \( q_e \) (mg/g) is the amount of dyes adsorbed at the equilibrium; \( Q_{\max} \) (mg/g) is the maximum adsorption capacity of the adsorbent; \( K_L \) (L/mg) is the Langmuir equilibrium constant; \( C_e \) (mg/L) is the dye concentration at the equilibrium.

Freundlich isotherm model:

\[
q_e = K_Fc_e^{1/n}
\]

(7)

where, \( K_F \) (mg/(g⋅(L/mg)]) is the Freundlich equilibrium constant; \( n \) is dimensionless exponent of the Freundlich equation.

Temkin isotherm:

\[
q_e = \frac{RT}{b} \ln(AC_e)
\]

(8)

It can be linearized as Eq. (9):

\[
q_e = B \ln A + B \ln C_e
\]

(9)

where, \( B = RT/b \), \( b \) (J/mol) is the Temkin constant related to heat of sorption, \( R \) (8.314 J/(mol-K)) is the gas constant and \( T \) (K) is the absolute temperature; \( A \) (L/g) is the Temkin isotherm constant.

2 Results and discussion

2.1 Preparation of CMC-AA adsorbent and factors on grafting efficiency of CMC-AA adsorbent

2.1.1 Weight ratio of AA to CMC

CMC-AA adsorbent is formed by free-radical chain polymerization. In free-radical polymerization, the weight ratio of AA and CMC (\( X_{AA} \)) markedly affects both the molecular weight of the polymer and GE (Said et al., 2004). During the reaction between AA and CMC, chain transfer and increase were directly correlated with the CMC quality. The viscosity of reaction system and steric hindrance effect also increased with gradually increased CMC weight. The effect of the \( X_{AA} \) on GE of CMC-AA adsorbent is shown in Fig. 1a. The GE of the product reaches a maximum of 82.3% at 10:3 of \( X_{AA} \) and then considerably decreases with increased CMC amount. The increase in GE with decreased weight ratio can be attributed to decreasing dissoluble homo-polymer of AA. Liu et al. (2009a) also considered that the overweight ratio of AA can be attributed to increased dissoluble homo-polymer of poly acrylate. With the further decreased \( X_{AA} \), the fraction of CMC increased, and the grafting of CMC and AA contributed to the increase in GE. A similar phenomenon was reported when CMC was used to synthesize CMC-based drug carrier (Ali et al., 2008).

2.1.2 Temperature

Figure 1b demonstrates the effect of the reaction temperature on GE of CMC-AA adsorbent. Increased temperature from 50°C to 70°C improves GE and the polymerization rate. The GE reached the maximum of 83.1% at 70°C and then decreased beyond this temperature. The diffusion rate of AA onto CMC increases at high temperature, resulting in a higher GE. In addition, the grafting reaction is chemically initiated by APS by producing radicals on the polymer chain. A more intense polymerization reaction occurs with increased temperature after chain initiation. However, APS is a thermo-sensitive initiator that does not need a very high temperature. Extreme temperatures also lead to many monomer groups grafting onto the main chain and forming a close network structure, thereby conferring difficulty to the application of APS (Ma et al., 2011).

2.1.3 pH values

Acrylic acid must be neutralized in the reaction process because its high activation at low pH produces a graft by itself. Once grafting AA it occurs, the grafting of CMC and AA is then inhibited. Figure 1c shows that the optimal pH for the grafting of CMC and AA is 7.0. After being neutralized with sodium hydroxide, the increasing hydrophilic groups advance the development of the network. The high pH inhibits the grafting reaction because of the repulsion of the negative charges of -COO\(^-\) groups. Highly
alkaline conditions are also reportedly disadvantageous to absorbent synthesis because more K⁺ or Na⁺ ions in the polymeric network at high pH values react with the -COO⁻ groups, resulting in the decrease of adsorption property of absorbent (Liu et al., 2009a; Ma et al., 2011).

2.1.4 Reaction time

Figure 1d indicates that GE increases with increased reaction time and reaches a maximum of 85.6% at 4 hr. The poly reaction processes of CMC and AA involve chain initiation, increase, termination and transfer. If the reaction time is too short, grafting is not completed. With prolonged reaction time, more three-dimensional network structures are formed, yielding to the gradually increased GE. At 4 hr, the free radical reaction is terminated following a constant GE. Similar results were obtained in grafting vinyl acetate onto starch (Samaha et al., 2005; Waly et al., 1998) and grafting AA onto guar gum (Taunk and Behari, 2000).

2.2 Characterization of CMC-AA adsorbent

The scanning electron microscopy images of CMC-AA adsorbent in Fig. 2b show a smooth surface with continuous phase but possessing pores. Pure CMC (Fig. 2a) is characterized with a rough surface. The pores in CMC-AA adsorbent can be one of the regions of water permeation and interaction sites with external stimuli in the graft.
polymers (Pourjavadi et al., 2010).

The Fourier-transform infrared spectroscopy (FT-IR) data of CMC-AA adsorbent (Fig. 3 line b) shows a new band at 1571 cm\(^{-1}\) (the stretching vibration of \(-\text{C}=\text{O}\)) following band reduction at 1607.9 cm\(^{-1}\) in CMC (Fig. 3 line a), which is due to grafting of \(\text{C}=\text{O}\) from AA (Said et al., 2004). The peaks at 1461 cm\(^{-1}\) and 1133 cm\(^{-1}\) in the spectrum of CMC-AA adsorbent (Fig. 3 line b) are related to the stretching vibration and symmetrical stretching of \(-\text{COO}\) groups (Pourjavadi et al., 2010). An absorption band at 622 cm\(^{-1}\) (Fig. 3 line b) formed because hydrogen bonding changed the position of the FTIR absorption bands from O-H stretching. All of analyses indicate the grafting reaction occurs between CMC and AA.

The porous characteristics of CMC-AA adsorbent are confirmed by nitrogen sorption tests. Table 1 shows that the Brunauer-Emmett-Teller (BET) surface areas of CMC-AA adsorbent are much higher than those of other adsorbent reported elsewhere (Silva et al., 2011). The obtained pore size distributions indicate a number of micropores with the diameters approximately in the range 4–8 nm (data not shown). Considering that the specific surface area and pore volume are the most important textural parameters to evaluate the adsorption capacity of an adsorbent (Haque et al., 2010), CMC-AA is considered as an alternative adsorbent for dye removal.

2.3 Adsorption of MO, DB and MG

2.3.1 Effect of initial dyes concentration on removal ratio

The adsorption capacities of dyes by CMC-AA adsorbent increased with increased dye concentration. However, the removal \((R)\) of dyes by CMC-AA adsorbent decreases when the concentration of the dye was increased to an inflection point (Fig. 4). When the concentration of MO, DB and MG was 30, 200 and 30 mg/L, respectively, the \(R\) of dyes is better than that at other experimental concentrations. Thus, these concentrations are adopted in subsequent experiments.

2.3.2 Effect of temperature and pH on the adsorption of MO, DB and MG

Dyes adsorption behavior on the adsorbents was investigated at different temperatures (Fig. 5a). With temperature increased from 25°C to 50°C, the \(R\) of CMC-AA adsorbent...
to increased MO, DB and MG also increased. However, the $R$ for MO, DB and MG becomes relatively constant beyond 35, 45 and 40°C, respectively. The increase in temperature may have promoted the permeation of dye molecule towards the absorbent because of swelling effect, but this increase can also lead to increased mobility of the dye ions (Wang et al., 2011).

The effects of the solution pH on the $R$ of dyes were studied by varying pH at 35, 45 and 40°C for the removal of MO, DB and MG, respectively. The results are shown in Fig. 5b. The optimal pH for the removal of DB is 7.0, growing the maximum removal of 79.6%. However, no peak removal is obtained for MO and MG efficient removal within the tested pH range. These values are replaced with the relatively constant $R$ of 84.2% and 99.9% at pH < 4.0 and > 9.0, respectively. $pH_{\text{PZC}}$ for CMC-AA adsorbent used is about 7.4. When the pH of MO dye solution is < 4.0, pH < $pH_{\text{PZC}}$, the CMC-AA adsorbent is positively charged. MO occurs in the anionic (pH < 3.1) or uncharged form (pH 3.1–4.4), the attraction forces between adsorbent surface and MO facilitates the MO adsorption. The high MO adsorption from wastewaters at low pH was proved to be due to neutralization of charge (Mittal et al., 2007). MG exists in an uncharged form, whereas CMC-AA adsorbent is negatively charged at solution pH > 11.0. Thus, the other absorption mechanisms of CMC-AA adsorbent to MG need further research. A high pH was also found for MG removal using activated slag as absorbent (Gupta et al., 1997). When it given the chemical structure and neutral pH property of DB, the similar charged state of CMC-AA adsorbent and DB at different pH values cause repulsion between the charged adsorbent and DB, resulting in a low $R$ of DB beyond the neutral pH range. A previous study has also verified that the neutral value is requisite for DB removal using CMC-AA adsorbent (Li et al., 2007).

pH affects the ionized state of an absorbent, the characteristics of dyes, and their interaction. Thus, preferable adsorption can be achieved by adjusting the pH of dyes solution. The three kinds of dyes in this study have difference molecular structure, but they can be efficiently removed at the respective pH condition by CMC-AA adsorbent. This result indicates that the universality of CMC-AA adsorbent in removing MO, DB and MG dyes.

All in all, the $R$ of MO, DB and MG also reached to 84.2%, 79.6% and 99.9% respectively under their optimized adsorption conditions, which are similar and even higher than previously reported (Haque et al., 2010; Fu et al., 2011; Khattri and Singh, 2009). It was found that the smaller the log $K_{\text{ow}}$ of dye is, the higher the $R$ is. Generally, if the compound has a low log $K_{\text{ow}}$ value (less than 10) it can be considered relatively hydrophilic. So the lower log $K_{\text{ow}}$ of MO, DB and MG (2.62, 3.50 and 0.62 respectively) may be an important factor for their high $R$ by CMC-AA adsorbent, a hydrophilic adsorbent with many -COO and -OH groups. In addition, based on the market price (USD 550–950) of activated carbon used removal of dyes, the cost that used CMC-AA adsorbent to adsorb dyes in a batch process was preliminarily calculated to be saved 10%–20%. Because the dyes are difficulty to be biodegraded, and the CMC-AA adsorbent is relative cheap and easy to be degraded, the spent adsorbent in a batch process will be treated by traditional chemical methods such as burning. Consequently, CMC-AA would be a good adsorbent for the removal of water-soluble dyes.

2.4 Kinetic studies

Both pseudo first-order and pseudo second-order kinetic models were used to fit the experimental data and to investigate the mechanism of adsorption and potential rate controlling steps such as mass transfer and chemical reaction. Figure 6 shows that the adsorption occurs through three stages for the different dyes. The first stage is rapid adsorption attributed to the rapid diffusion and adsorption of the dye to CMC-AA adsorbent surface (Silva et al., 2011). The second stage is a delayed process due to the intraparticle diffusion and is close to adsorption...
equilibrium. The third stage is equilibrium establishment. The highest \( q_e \) value is found for DB adsorption onto CMC-AA adsorbent, followed by MG and DO.

Table 2 shows that the \( R^2 \) values range from 0.7306 to 0.8882 for the intraparticle diffusion model. If adsorption process follows the intraparticle diffusion process, a plot of \( q_t \) against \( t^{1/2} \) should be a straight line (Bekci et al., 2009). Generally, adsorption process does not follow intraparticle diffusion process, the adsorption is less affected by intraparticle diffusion. Thus, the result shows that the DB, MG and MO adsorption by CMC-AA adsorbent is less affected by intraparticle diffusion. By contrast, the pseudo-second-order model presents the best fit, presenting high \( R^2 \) values (\( R^2 > 0.99 \)) for three kinds of dyes, and the calculated \( q_e \) agreed with the experimental data. According to this model, the adsorption rate depends on the amount of dye adsorbed on the surface of CMC-AA absorbent and the amount adsorbed at equilibrium. The overall rate of the dye adsorption process should be controlled by chemical adsorption. Similar phenomena have been observed for DB adsorption (Özacar and Sengil, 2004).

So far many adsorbents have been evaluated as candidates for the removal of MO from water and their adsorption capacities have varied widely depending on the adsorbent. Even though the adsorption capacity of CMC-AA is less than that of iron terephthalate and activated carbon (Haque et al., 2011), its capacity is relatively greater than that of the most other adsorbents (Haque et al., 2010; Liu et al., 2009b). The maximum adsorption capacity of CMC-AA adsorbents for MG (147.9 mg/g) is the highest among celllose-based and starch-based absorbent materials such as rattan sawdust (Hameed and El-Khaiary, 2008) and cross linked amphoteric starch (Xu et al., 2006). Likewise, adsorption capacity of CMC-AA adsorbents for DB was the highest among previously reported values (Fu et al., 2011).

### 2.5 Equilibrium models

Adsorption isotherms describe how adsorbates interact with adsorbents and are critical in optimizing the use of adsorbents. In this study, the Langmuir isotherm, Freundlich isotherm and Temkin isotherm were used for describing the results. Table 3 shows that the Langmuir isotherm and Freundlich isotherms poorly suit to the adsorption of DB and MG in comparison to Temkin isotherm. The validity of the Temkin model suggests that the DB and MG dyes uptakes have a uniform distribution of binding energies. Thus, the heat of adsorption of all the molecules in the

<table>
<thead>
<tr>
<th>Sample</th>
<th>CMC-AA</th>
<th>Langmuir isotherm</th>
<th>Freundlich isotherm</th>
<th>Temkin isotherm</th>
</tr>
</thead>
<tbody>
<tr>
<td>MO</td>
<td>1111.11</td>
<td>0.007</td>
<td>7.82</td>
<td>2.94</td>
</tr>
<tr>
<td>DB</td>
<td>1096.05</td>
<td>0.001</td>
<td>20.70</td>
<td>21.98</td>
</tr>
<tr>
<td>MG</td>
<td>555.56</td>
<td>0.021</td>
<td>10.37</td>
<td>2.36</td>
</tr>
</tbody>
</table>

The values were obtained by varying the initial concentration of MO, DB and MG under the adsorption conditions of pH 5.0, 7.0, 9.0, temperature of 35, 45 and 40°C respectively, contact time 2 hr, and CM-AA 0.4 g/L.
layer decreased linearly with coverage due to adsorbent-adsorbate interactions (Deng et al., 2009). The high $R^2$ of Freundlich and Temkin isotherm are observed in MO adsorption, suggesting that several interactions exist between CMC-AA adsorbent and MO.

3 Conclusions

A new cellulose-based absorbent CMC-AA was prepared by grafting polymerization of AA onto CMC. The highest GE value of 85.6% is obtained under the optimum synthesis conditions. The prepared CMC-AA absorbent is proved to be a potential adsorbent for removal of MO, DB and MG from aqueous solutions, although the adsorption depends on different temperatures and pH values. Pseudo second-order kinetic model better fits the kinetics of current adsorption, showing the chemical adsorption of CMC-AA adsorbent to MO, DB and MG. Temkin isotherm better fits the experimental equilibrium data of dye adsorption on the prepared CMC-AA absorbent.

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