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Remediation capacity of carboxymethyl lignocellulose/montmorillonite composite materials for monosodium glutamate wastewater

Baiyun Zhao†, Ze Liu†, Yanrong Liu, Qi Shi, Xiaokai Liu, Li Wang*

College of Material Science and Art Design, Inner Mongolia Agricultural University, Laboratory of Fibrosis and Energy Utilization of Shrubby Resources in Inner Mongolia Autonomous Region, Hohhot 010018, China, emails: wl2083663@126.com/wlndcl@126.com (L. Wang), 1425491996@qq.com (B. Zhao), 2823668765@qq.com (Z. Liu), 811273066@qq.com (Y. Liu), 996713669@qq.com (Q. Shi), 779960624@qq.com (X. Liu)

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ABSTRACT

Carboxymethyl lignocellulose/montmorillonite composite (CNC/MMT) was prepared by intercalation-polymerization with carboxymethyl lignocellulose (CNC) and montmorillonite (MMT). When the mass ratio of carboxymethyl lignocellulose and montmorillonite of 0.5:1, preparation time of 6 h, and preparation temperature of 60°C, the declining values after treatment of chemical oxygen demand (COD) and ammonia nitrogen (NH_3–N) in monosodium glutamate wastewater (MSGW) by CNC/MMT are 8,786 and 3,700 mg/L, respectively. The carboxymethyl lignocellulose/montmorillonite was characterized by scanning electron microscopy, transmission electron microscopy, Brunauer–Emmett–Teller, Fourier-transform infrared spectroscopy, X-ray diffraction, and thermogravimetric analysis. The results showed that the –COO– in carboxymethyl lignocellulose and Si–OH in montmorillonite form CNC/MMT with rough surface through complexation and coordination, and the crystal structure of montmorillonite is destroyed, forming intercalation-exfoliation carboxymethyl lignocellulose/montmorillonite. The effects of carboxymethyl lignocellulose/montmorillonite on the removal capacity of COD and NH_3–N in MSGW were investigated by changing the experimental conditions. When the mass of carboxymethyl lignocellulose/montmorillonite of 0.1 g, the contact temperature of 60°C, and contact time of 6 h, the declining values of COD and NH_3–N are 8,979 and 3,884 mg/L, respectively.

Keywords: Carboxymethyl lignocellulose (CNC); Monosodium glutamate waster (MSGW); Montmorillonite (MMT); Natural wastes

1. Introduction

Monosodium glutamate (MSG), as a food additive, is a common condiment on people's table. The appropriate capacity of adding MSG will make dishes bright colour and taste delicious. However, a large amount of monosodium glutamate wastewater (MSGW) is produced while enterprises produce more MSG to meet people's demands. MSGW is a kind of organic wastewater of high concentration, treatment difficulty and the treatment process is complex. If directly discharged to the environment, monosodium glutamate wastewater will cause bioaccumulation, leading the water body to turn black and stink, thus polluting the environment and affecting the tissues of living beings. Therefore, it is particularly important to explore more efficient treatment methods to reduce water pollution.

At present, a variety of wastewater treatment technologies have appeared at home and abroad. Among them, the adsorption method [1,2] has the advantages of simplicity, effectiveness, low cost and easy operation as compared with other conventional methods (such as precipitation). Especially in the aspects of economy and environmental protection, this method can effectively reduce the cost and secondary pollution to the environment. Biomass adsorption

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* Corresponding author.

† Both the authors contributed equally to this paper.

technology is an emerging technology [3,4], which utilizes natural wastes to adsorb pollutants from industrial wastewater. Natural waste is abundant, widely available, and inexpensive. Therefore, natural waste can be used as an adsorbent for wastewater treatment.

Lignocellulose (LNC) [5–8] is mainly composed of carbohydrate polymers (cellulose and hemicellulose) and aromatic macromolecules (lignin). LNC has attracted more and more attention in wastewater treatment due to its good adsorption and biodegradability. It has the advantages of low cost, no-toxicity, tasteless, large specific surface area [9], rich chemical bonds, high cellulose content, and so on. So far, it has been applied to the treatment of a variety of organic wastewater [10,11], and the treatment mechanisms mainly include Van der Waals force, surface complexation and ion exchange. It can be seen that LNC has good adsorption performance. In addition, it can have more carboxyl hydrophilic groups through chemical modification, and the wastewater treatment performance can be further improved. In our previous study [11], LNC has been modified with carboxymethyl groups to combine –COO– into the material, which improved the adsorption effect of Congo red.

As a layered silicate clay mineral, montmorillonite (MMT) has the advantages of large specific surface area and large cation exchange capacity. Therefore, it has a good adsorption performance and is widely used in the treatment of wastewater [12–14]. Up to now, researchers have intercalated montmorillonite into chitosan to obtain not only a high cationic dye treatment performance but also enhanced its thermal stability by filling MMT in the polymer system [15]. When montmorillonite alone is used as treatment agent, the treatment effect is poorer. Therefore, it is urgent to find an environmentally friendly treatment agent (such as biopolymer and clay mineral) for efficiently treating wastewater.

In view of the shortcomings of carboxymethyl lignocellulose (CNC) and montmorillonite as adsorbents for wastewater treatment, there are few reports about their application in MSGW treatment. CNC/MMT is prepared by using carboxymethyl lignocellulose and montmorillonite, which not only bring the advantages of both into play, but also make up for their shortcomings. The CNC/MMT is used for treating monosodium glutamate wastewater and has the advantages of simple operation, short process flow, cost saving and easy separation and recovery, and provides theoretical basis for the treatment of MSGW.

Table 1
Specific surface area and average pore size of CNC, MMT and CNC/MMT

Samples	Specific surface area ($m^2 \cdot g^{-1}$)	Average pore size (nm)
CNC	1.07	11.98
MMT	60.12	9.56
CNC/MMT	27.04	8.99

2. Materials and methods

2.1. Materials and instruments

MSGW was provided by Inner Mongolia Fufeng Biological Technology Co., Ltd., China. The chemical oxygen demand (COD) concentration of 80,000–90,000 mg/L, ammonia nitrogen (NH_3–N) concentration of 26,000–30,000 mg/L, pH of 3.4, and the colour is dark black.

LNC was produced in Beijing Qinli Hengtong Co., Ltd., China. Montmorillonite was produced in Xinglong Binder Chemical Co., Ltd., China. Sodium hydroxide (NaOH), acetic acid (CH_3COOH), anhydrous ethanol (CH_3CH_2OH) and chloroacetic acid ($ClCH_2COOH$) were produced in Fuchen Chemical Reagent Co., Ltd., Tianjin, China. C1, C2 (special reagent for COD testing) and N1, N2 (special reagent for NH_3–N testing) were produced in Jiangsu Shengaohua Environmental Protection Technology Co., Ltd., China.

Vacuum drying oven (DZF-6210; Jintan Jingda Instrument Manufacturing Co., Ltd., China). Digital display water bath constant temperature oscillator (SHA-C; Jintan City Jiangnan Instrument Factory, China). Centrifuge (H2050R; Changsha Xiang Instrument Centrifuge Instrument Co., Ltd., China). Circulating water vacuum pump (SHZ-III; Shanghai Ya Rong Biochemical Instrument Factory, China). pH meter (STARTER; Auhaus Instruments (Shanghai) Co., Ltd.).

2.2. Synthesis of carboxymethyl lignocellulose

The synthesis process of carboxymethyl lignocellulose is as shown in Fig. 1. 10 g of LNC was weighed, and added into 100 mL of 20 wt.% NaOH solution, mixed under magnetic stirring for 30 min and immersed for 12 h. After filtration and drying, the sample was transferred to a three-necked flask, and 100 mL CH_3CH_2OH was added. Then, 8 g of $ClCH_2COOH$ was slowly added into three batches, reacted at room temperature for 30 min, heated to 60°C for 6 h, and filtered to obtain a product. The product was added

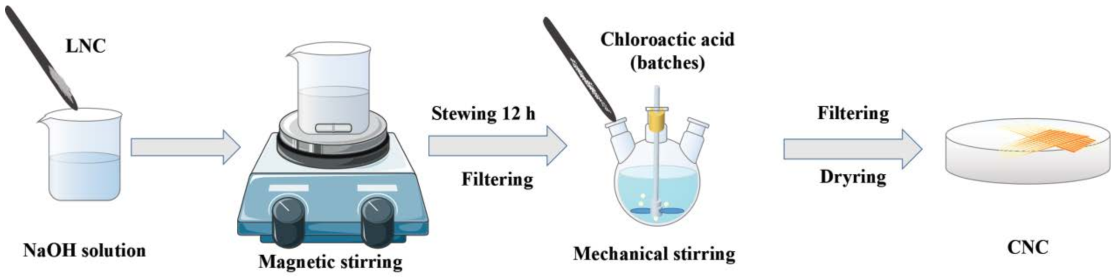

Fig. 1. Synthesis process of CNC.

in 50 mL distilled water, adjusted to neutrality with 50 wt.% CH_3COOH or NaOH, after filtered, dried at 80°C for 4 h, and ground to obtain CNC.

2.3. Synthesis of CNC/MMT

The synthesis process of CNC/MMT is as shown in Fig. 2. 4 g of carboxymethyl lignocellulose was added into 120 mL distilled water (the ratio of carboxymethyl lignocellulose (g) to distilled water (mL) was 1:30), and the homogeneous suspension was formed by magnetic stirring at 25°C for 30 min. Montmorillonite suspension (1 g montmorillonite was added into 30 mL of distilled water by magnetic stirring at 25°C for 30 min) and carboxymethyl lignocellulose suspension was slowly added into a three-necked flask, heated at 60°C for 6 h. Then, the pH was adjusted to be neutral with 50 wt.% CH_3COOH or NaOH. Finally, the product was washed with distilled water, filtered, dried at 80°C for 4 h and ground to obtain carboxymethyl lignocellulose/montmorillonite.

2.4. Treatment experiments of MSGW with CNC/MMT

Firstly, 25 mL of MSGW and 0.10 g CNC/MMT were added to a conical flask with stopper and medicine spoon. Then, the conical flask was put into a constant temperature water bath to oscillate. MSGW was treated under different conditions (the mass of CNC/MMT, contact temperature and contact time). Finally, the samples were centrifuged and the supernatants were taken to determine their COD and NH_3–N concentration.

2.5. Determination of COD and NH_3–N concentration

Firstly, 3 mL of MSGW was added to a sealed tube. Then, 1 mL of C1 and 5 mL of C2 were added in sequence, shake evenly by hand, the tube was digested in a digester at 168°C for 10 min. Afterwards, the tube was cooled to room temperature, 3 mL of distilled water was added, shaken evenly by hand. The value of COD in MSGW was measured by using the rapid digestion spectrophotometry (6B-200, Jiangsu Shengaohua Environmental Protection Technology Co., Ltd., China). The average value of the three groups of data was measured. The determination principle is special reagents (C1 and C2) are used as the oxidant and catalyst, reducing Cr(VI) to Cr(III), the value of COD is determined by colorimetric method.

Firstly, 10 mL of MSGW, 1 mL N1 and 1 mL N2 were added in the tube, shake evenly by hand, the tube was held in a rack for 10 min. The concentration of NH_3–N in the solution was measured by using the Nessler's colorimetric method (6B-50, Jiangsu Shengaohua Environmental Protection Technology Co., Ltd., China). The average value of the three groups of data was measured. The determination principle is to reduce nitrite in solution into ammonia.

2.6. Characterization

The surface microstructure of the sample was analysed by a scanning electron microscope (SEM; Hitachi S-4800, Japan Hitachi Company). The transmission electron microscope (TEM; Talos F200C G2, Thermo Fisher Scientific Shier Technology) was used for observing the microstructure of the composite material. The nitrogen adsorption–desorption method was used to determine the specific surface area and average pore diameter of the samples using the pore specific surface area analyzer (BET; ASAP 2460, Mike Instruments Company of America). The Fourier-infrared spectrometer (FT-IR; Tensor27, Bruker Company of Germany) was an instrument for measuring the change of the functional group of the sample. The samples were treated with KBr, and the scanning ranges from 400 to 4,000 cm^{-1}. X-ray diffractometer (XRD; PANalytical, Netherlands PANalytical Company) can measure the crystal structure and phase composition of samples. The samples were placed on a blank slide with an incident wavelength of 0.514 nm, scanning ranges 10°–90°. The thermal stability of materials was determined by thermogravimetric analyzer (TG; STA409PC, Netzsch Company of Germany).

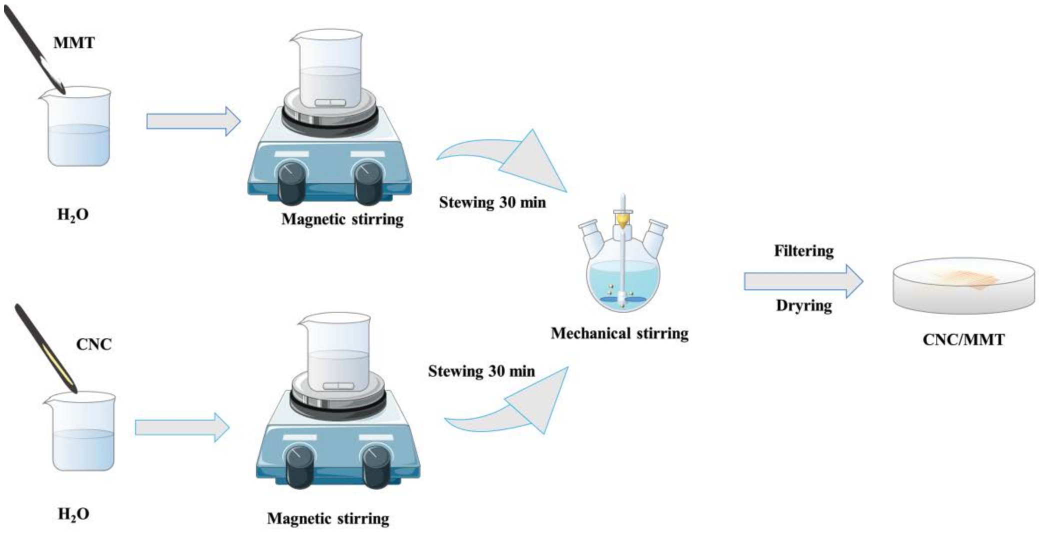

Fig. 2. Synthesis process of CNC/MMT.

3. Results and discussion

3.1. Effects of the preparation conditions for remediation capacity of MSGW by CNC/MMT

3.1.1. Mass ratio of CNC and MMT

Fig. 3a shows the declining values after treatment of COD and NH_3–N in MSGW by CNC/MMT or carboxymethyl lignocellulose and montmorillonite (CNC/MMT) first increased and then decreased with the increase of the mass ratio of carboxymethyl lignocellulose and montmorillonite. When the content of carboxymethyl lignocellulose is less, montmorillonite slice layer cannot be saturated filling. With the increase of carboxymethyl lignocellulose content, the layers of the montmorillonite slices have been saturated filled [16], thus improving the removal capacity of CNC/MMT for COD and NH_3–N. The declining values of COD and NH_3–N by carboxymethyl lignocellulose/montmorillonite are 8,786 and 3,700 mg/L, respectively. When the content of carboxymethyl lignocellulose is too much, the bonding degree between carboxymethyl lignocellulose and montmorillonite molecules will increase, resulting in the decrease of voids in the composites, which is not conducive to the treatment of MSGW by CNC/MMT.

3.1.2. Preparation temperature of CNC/MMT

Fig. 3b shows the declining values after treatment of COD and NH_3–N in MSGW by CNC/MMT first increased and then decreased with the rise of preparation temperature. The reaction activity is low at low temperature, which is not conducive to the intercalation reaction of montmorillonite and carboxymethyl lignocellulose. With the rise of temperature, the reaction activity and collision probability of carboxymethyl lignocellulose and montmorillonite increased. The hydrogen bonds between carboxymethyl lignocellulose molecules are fractured into small molecules, which is beneficial to the opening of montmorillonite slices and the entry of carboxymethyl lignocellulose small molecules. However, if the temperature is too high, CNC is easily degraded, which aggravates the intermolecular movement and relatively reduces the binding opportunity of carboxymethyl lignocellulose and the montmorillonite layer, thereby reducing the declining values of CNC/MMT for COD and NH_3–N in monosodium glutamate wastewater.

3.1.3. Preparation time of CNC/MMT

Fig. 3c shows that the declining values after treatment of COD and NH_3–N in MSGW by CNC/MMT first increased and then decreased with the prolonging of preparation time. At the initial stage of the reaction, there are many gaps between montmorillonite sheets and more active sites on the carboxymethyl lignocellulose surface, which promoted the intercalation reaction. With the prolongation of time, the interlayer of montmorillonite sheets are gradually filled saturated, and the removal capacity of carboxymethyl lignocellulose/montmorillonite for COD and NH_3–N in monosodium glutamate wastewater is gradually improved. Subsequently,

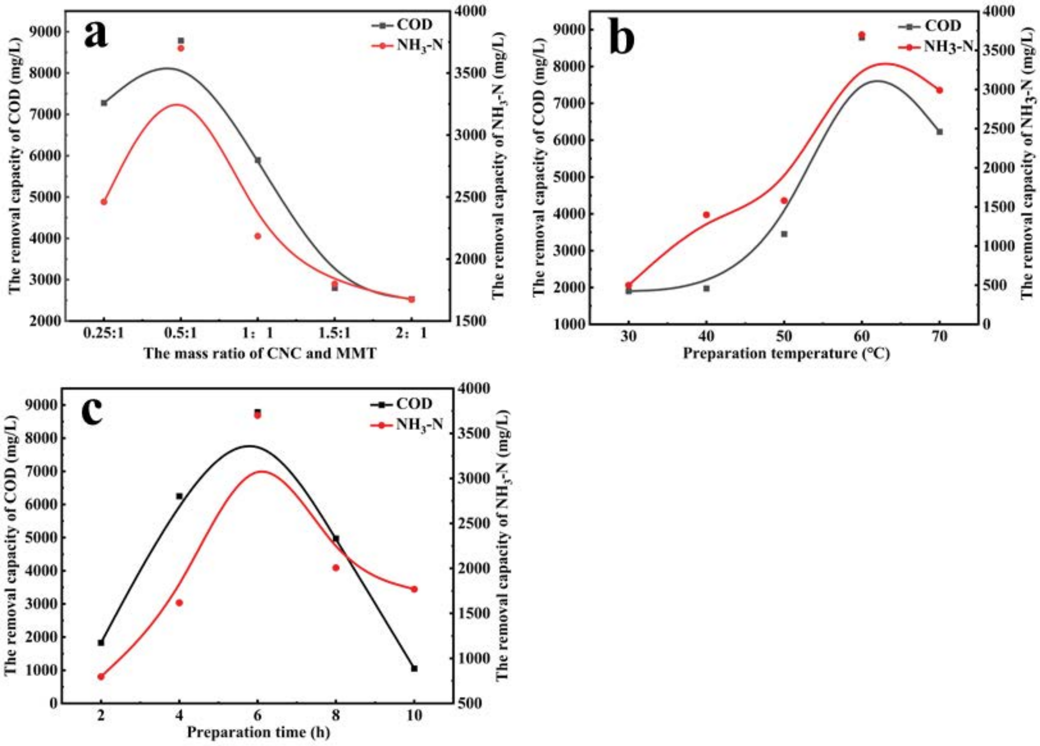

Fig. 3. Effects of mass ratio of CNC and MMT (a), preparation temperature (b), and preparation time (c) of CNC/MMT on removal capacity of COD and NH_3–N in MSGW (Treatment condition: 0.1 g CNC/MMT treating 25 mL MSGW; treatment temperature of 60°C; time of 6 h).

the preparation time exceeds 6 h, and the un-intercalated carboxymethyl lignocellulose molecules can twine with each other to destroy the stability of the system, which is not conducive to the dispersion of montmorillonite in solution, thus reducing the declining values of CNC/MMT for COD and NH_3–N in MSGW.

3.2. Characterization results

3.2.1. Scanning electron microscopy

The micromorphology of the montmorillonite (a) and CNC/MMT (b) were studied by SEM (Fig. 4a and b). Fig. 4b shows that the smooth surface of montmorillonite appears the irregular and dispersed lamination stack in the shape of curl or lump, which indicates that intercalation reaction occurs between carboxymethyl lignocellulose and montmorillonite, and the pores in the structure increase. These pores increase the contact area, which is beneficial to the combination of carboxymethyl lignocellulose/montmorillonite with organic substances in wastewater, and improves the removal capacity of carboxymethyl lignocellulose/montmorillonite for COD and NH_3–N in MSGW.

3.2.2. Transmission electron microscopy

The macrostructure of the carboxymethyl lignocellulose/montmorillonite was studied by TEM (Fig. 4c). The black strips and clusters are peeled off montmorillonite sheets, and the white materials are carboxymethyl lignocellulose materials that entered the montmorillonite interlayer structure. Furthermore, carboxymethyl lignocellulose has entered the montmorillonite layer, which shows that a relatively stable intercalation-peeling composite material (CNC/MMT) is formed.

3.2.3. Brunauer–Emmett–Teller

Total area per gram of sample and average pore size of the sample (specific pore volume: surface area ratio) were tested by Brunauer–Emmett–Teller. The specific surface area of carboxymethyl lignocellulose/montmorillonite (27.04 $m^2\cdot g^{-1}$) is smaller than montmorillonite (60.12 $m^2\cdot g^{-1}$), and is larger than carboxymethyl lignocellulose (1.07 $m^2\cdot g^{-1}$). This indicates that the specific surface area of CNC and MMT is improved, because most of the holes of the montmorillonite are occupied by the carboxymethyl lignocellulose. The average pore size of CNC/MMT (8.99 nm) is smaller than that of carboxymethyl lignocellulose (11.98 nm) and montmorillonite (9.56 nm), thereby improving the declining values of carboxymethyl lignocellulose/montmorillonite for COD and NH_3–N in MSGW.

3.2.4. Fourier-transform infrared spectroscopy

The functional groups of carboxymethyl lignocellulose, montmorillonite and carboxymethyl lignocellulose/montmorillonite were studied by FT-IR (Fig. 5a). The characteristic

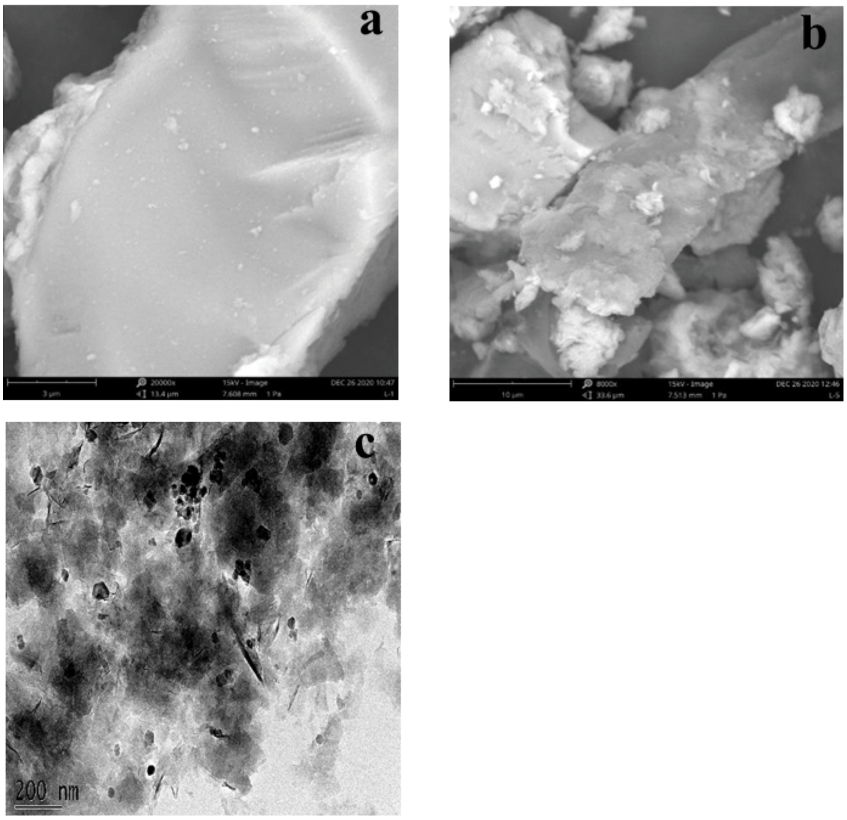

Fig. 4. SEM images of MMT (a) and CNC/MMT (b), and TEM image of CNC/MMT (c).

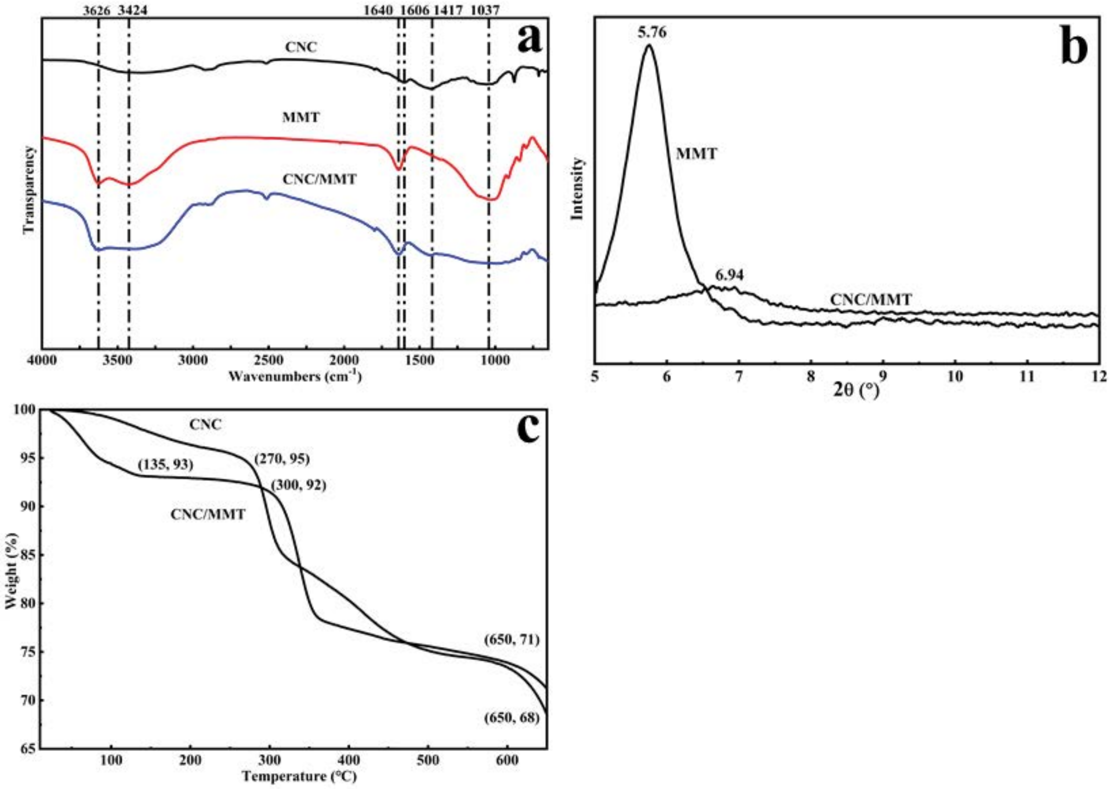

Fig. 5. FT-IR (a) spectra of CNC, MMT and CNC/MMT, XRD (b) spectra of MMT and CNC/MMT, and TG (c) spectra of CNC and CNC/MMT.

peaks of carboxymethyl lignocellulose corresponding to –COO– are located at 1,606 and 1,417 cm^{-1}, and the C=C peak at 1,640 cm^{-1} [17]. The characteristic peaks of montmorillonite corresponding to Si–O, Si–OH and O–H stretching vibration absorption peak of H_2O are located at 1,037, 3,626 and 3,424 cm^{-1} [18]. The intensity of characteristic peaks of the CNC/MMT are weakened at 1,037 and 3,626 cm^{-1}, O–H of H_2O in montmorillonite disappeared at 3,424 cm^{-1} due to the form of intermolecular hydrogen bonds, C=C in montmorillonite at 1,640 cm^{-1} and –COO– in carboxymethyl lignocellulose at 1,606 and 1,417 cm^{-1} are weakened. Therefore, the –COO– bond in carboxymethyl lignocellulose and the Si–O, Si–OH bond in montmorillonite are coordinated or complexed to form carboxymethyl lignocellulose/montmorillonite.

3.2.5. X-ray diffraction

The crystal structure of the montmorillonite and CNC/MMT were studied by XRD (Fig. 5b) [19]. The characteristic diffraction peak of montmorillonite appears at the diffraction angle 2θ of 5.76° (110). According to the Bragg equation ($2d\sin\theta = k\lambda$ (k = 1, 2, 3 …), the interlayer spacing of montmorillonite d = 1.49 nm. The diffraction peak (110) of MMT in carboxymethyl lignocellulose/montmorillonite obviously weakened and shifted to a large angle (6.94°), which indicated that carboxymethyl lignocellulose destroyed the crystal structure of montmorillonite. The montmorillonite layers are occupied to form the CNC/MMT.

3.2.6. Thermogravimetric analysis

The thermostability of the carboxymethyl lignocellulose and carboxymethyl lignocellulose/montmorillonite were studied by TG (Fig. 5c). The weight loss rate of carboxymethyl lignocellulose is 5% from room temperature to 270°C due to the loss of free water. The obvious weight loss from 270°C to 650°C (weight loss rate is 32%), mainly due to the depolymerisation of carboxymethyl lignocellulose. The weight loss rate of CNC/MMT is 7% from room temperature to 135°C, which may be due to the evaporation of water molecules and the decomposition of impurities. The apparent weight loss occurs from 300°C to 650°C (weight loss rate is 29%). These indicate that a slight increase in the thermal stability of the carboxymethyl lignocellulose/montmorillonite relative to the carboxymethyl lignocellulose.

3.3. Effects of the treatment conditions for remediation capacity of MSGW by CNC/MMT

3.3.1. Mass of CNC/MMT

Fig. 6a shows the declining values after treatment of COD and NH_3–N in MSGW by CNC/MMT first increased and then gradually levelled off with the increase of carboxymethyl lignocellulose/montmorillonite mass. The declining values of COD and NH_3–N in monosodium glutamate wastewater by carboxymethyl lignocellulose/montmorillonite are 8,979 and 3,884 mg/L, respectively. The organic substances in

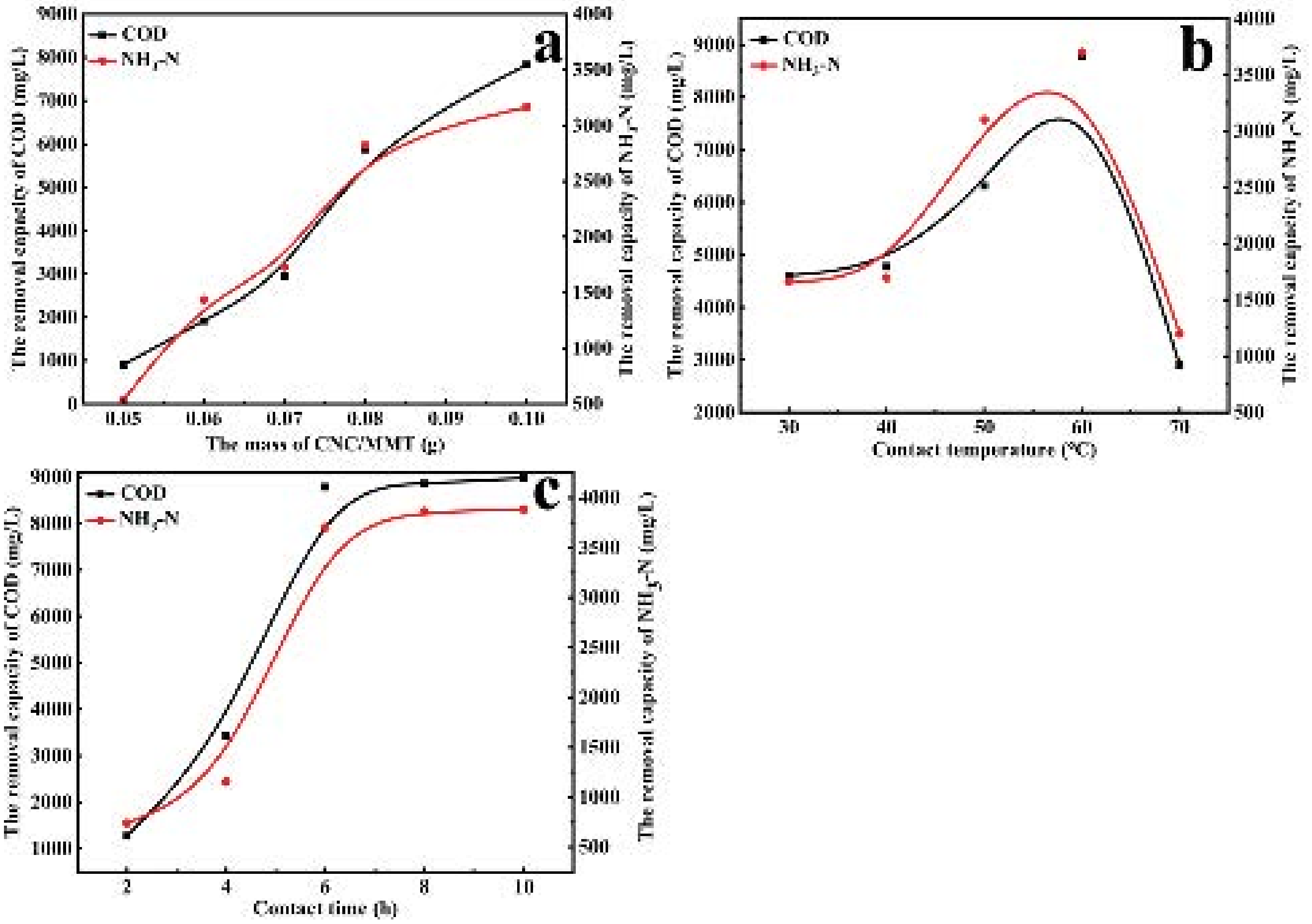

Fig. 6. Effects of the mass of CNC/MMT (a), contact temperature (b) and contact time (c) on removal capacity of COD and NH_3–N in MSGW (Preparation condition: the mass ratio of CNC and MMT of 0.5:1; preparation temperature of 60°C; preparation time of 6 h).

monosodium glutamate wastewater are adsorbed by CNC/MMT, gathered on the surface of carboxymethyl lignocellulose/montmorillonite, and finally settled together with carboxymethyl lignocellulose/montmorillonite [20], thus reducing the declining values of carboxymethyl lignocellulose/montmorillonite for COD and NH_3–N in MSGW.

3.3.2. Contact temperature

Fig. 6b shows the declining values after treatment of COD and NH_3–N in MSGW by CNC/MMT first increased and then decreased with the rise of contact temperature. With the rise of the contact temperature, the molecular thermal motion leads to the increase of the carboxymethyl lignocellulose/montmorillonite surface active groups, and the more likely for the organics in monosodium glutamate wastewater to contact with CNC/MMT, thus increasing the declining values of CNC/MMT for COD and NH_3–N in monosodium glutamate wastewater. With the further rise of temperature, the thermal motion accelerated, causing the increase of the irregular collision in the system, and the organic substances in the monosodium glutamate wastewater did not have enough time to combine with the carboxymethyl lignocellulose/montmorillonite, thus reducing the declining values of carboxymethyl lignocellulose/montmorillonite for COD and NH_3–N in MSGW.

3.3.3. Contact time

Fig. 6c shows the declining values after treatment of COD and NH_3–N in MSGW by CNC/MMT first increased and then gradually levelled off with the prolonging of contact time. Firstly, the organic substances had insufficient contact with the carboxymethyl lignocellulose/montmorillonite, and low declining values due to the short contact time. With the prolongation of time, CNC/MMT absorbed more organic substances and the declining values reached the balance after 6 h.

4. Conclusions

The preparation results showed that when the mass ratio of carboxymethyl lignocellulose and montmorillonite is 0.5: 1, the preparation temperature and time are 60°C and 6 h. The declining values after treatment of COD and NH_3–N in MSGW by CNC/MMT are 8,786 and 3,700 mg/L, respectively.

The characterization results show that the –COO– in carboxymethyl lignocellulose and Si–OH in MMT form carboxymethyl lignocellulose/montmorillonite with rough surface through complexation and coordination. The introduction of carboxymethyl lignocellulose destroyed the crystal structure of montmorillonite, thus forming the CNC/MMT, at the same time also increasing thermal stability. The surface of the compounded material is rough and porous, which is more favourable for the adsorption reaction.

The treatment results showed that treatment MSGW with 0.1 g carboxymethyl lignocellulose/montmorillonite for 25 mL, treatment at 60°C for 6 h. The declining values after treatment of COD and NH_3–N in monosodium glutamate wastewater by CNC/MMT are 8,979 and 3,884 mg/L, respectively. The CNC/MMT has good removal capacity for COD and NH_3–N, and has excellent performance including environmental protection, economy and regeneration, which provides a certain theoretical basis for treatment of COD and NH_3–N in MSGW. At the same time, it also provides a theoretical guidance for the actual wastewater treatment.

Acknowledgments

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