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Poly(Amic Acid)-Modified Biomass of Baker's Yeast for Enhancement Adsorption of Methylene Blue and Basic Magenta

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Abstract In this study, poly(amic acid)-modified biomass was prepared to improve the adsorption capacities for two cationic dyes, methylene blue and basic magenta. X-ray photoelectron spectroscopy and potentiometric titration demonstrated that a large number of imide, amine, and carboxyl groups were introduced on the biomass surface, and the concentrations of these functional groups were calculated to be 0.27, 1.08, and 1.08 mmol g^{-1} by using the first derivative method. According to the Langmuir equation, the maximum uptake capacities (q_m) for methylene blue and basic magenta were 680.3 and 353.4 mg g^{-1}, respectively, which were 13- and sevenfold than that obtained on the unmodified biomass. Adsorption kinetics study showed that the completion of the adsorption process needed only 40 min, which is faster than the common sorbent such as activated carbon and resin. Experimental results showed that pH and ionic strength had little effect on the capacity of the modified biomass, indicating that the modified biomass had good potential for practical use.

Keywords Modified biomass · Methylene blue · Basic magenta · Adsorption · First derivative method

Introduction

Industrial activity is responsible for generating a large volume of hazardous effluents. Color is one of the most important hazard effluents in industrial effluents, which needs to be treated [1–2] because the presence of dyes in water reduces light penetration, precluding the photosynthesis of aqueous flora [3–4]. Besides that, some dyes may do harm to humans [5]. The majority of technologies presently employed for color removal are based on

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physicochemical processes such as dilution, adsorption, coagulation and flocculation, chemical precipitation, oxidation, ion exchange, reverse osmosis, and ultrafiltration [6, 7]. Of all these procedures for the removal of synthetic dyes from industrial effluents, the adsorption procedure is the most efficient because the hazardous species is transferred from the water effluent to a solid phase, diminishing the effluent volume to a minimum [4, 8, 9]. The adsorbent can be regenerated afterwards or kept in a dry place without direct contact with the environment [10]. But the price of the common adsorbent such as activated carbon and resin is high, limiting its large-scale application in wastewater treatment. Therefore, there is a growing interest in finding alternative low-cost adsorbents for dye removal from aqueous solution.

Recently, more and more attention was paid on biosorbents due to their low cost. Literature had reported that a wide variety of microorganisms, including bacteria, algae, and fungi, were capable of sorbing a number of pollutants [11–14]. Among the microorganisms used for biosorption, baker's yeast is an inexpensive, readily available source of biomass for dye removal from wastewater. Cell walls of fungal biomass including that of baker's yeast contain chitin, chitosan, β-1,3-D-glucans, β-1,6-D-glucans, and mannoproteins which are sources of different functional groups such as carboxyl, amine, hydroxyl, phosphate, and sulfonate [15]. These functional groups have been reported to be responsible for cationic dye binding [16]. However, the density of these functional groups effective for adsorption is generally low, and most biosorbents do not show a high sorption capacity for cationic dyes [16, 17].

Since the adsorption of dyes takes place mainly on the surface, increasing the density of the functional groups on the biomass would be an effective approach to enhance the adsorption capacity. It had been reported that the adsorption capacity of the biosorbent for metal ions could be improved greatly through chemical modification [18–21].

Poly(amic acid) from reaction of pyromellitic dianhydride (PMDA) and lysine (Fig. 1) is composed of a large number of carboxyl and amine groups in a molecule. It exhibits good adsorption ability for metal ions and cationic dyes. The adsorption of baker's yeast for cationic dyes would increase significantly after modification with poly (amic acid).

In this study, the biomass of baker's yeast was modified with poly(amic acid) through a chemical modification method. The modified biomass was characterized by X-ray photoelectron spectroscopy (XPS) and transmission electron microscopy (TEM). The amounts of the functional groups including carboxyl, amine, and imide groups on the modified biomass were determined by potentiometric titration curve using the first derivative method. The adsorption performances for the two common dyes, methylene blue and basic magenta, were studied, and the effects of pH, temperature, and ionic strength on the adsorption were also investigated.

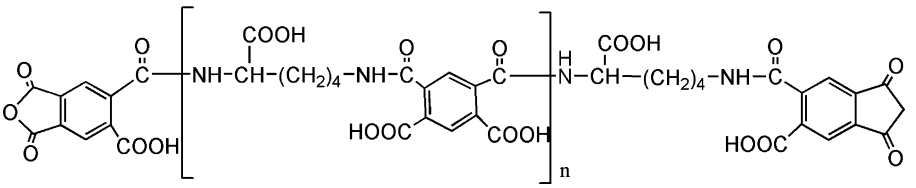

Fig. 1 Structure of poly(amic acid)

Materials and Methods

Materials

PMDA and lysine were purchased from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). The two dyes, methylene blue and basic magenta, were purchased from Ryond Chemical Co., Ltd, and they were used without further purification. Their structures were shown in Fig. 2. Baker's yeast was purchased from China General Microbiological Culture Center (Beijing, China) and it was dried at 60 °C for 24 h before use. Other chemicals were reagent grade.

Surface Modification

The modified biomass was prepared through chemical grafting method [22]. Added to a 30-mL *N, N*-dimethylacetamide (DMAc) were 1.0 g of PMDA and 0.2 g of lysine. After stirring at room temperature for 2 h, poly(amic acid) was formed. Then, 0.5 g of the biomass was added and continuously stirred for another 4 h at 50 °C. The obtained biomass was washed in the following order with DMAc, NaOH (0.1 mol L^{-1}), and distilled water to remove residual monomer and polymer and then freeze-dried and stored in a desiccator before use.

Potentiometric Titration

Before the titration, 0.05 g of the biomass was added to conical flasks containing 20 mL of 0.01 mol L^{-1} NaCl solution, and the mixture was allowed to stand for 12 h at room temperature for stabilization before the solution was bubbled with nitrogen gas for 2 h with vigorous mixing to remove carbon dioxide in the solution [23–25]. Titration was performed on a titrator (ZDJ-400, China) using 0.1501 mol L^{-1} HCl solution. The amount of HCl consumed and the solution pH were recorded after equilibrium was attained.

Batch Adsorption Experiments

Batch adsorption experiments were conducted at 25 °C and 150 rpm on an orbital shaker. A 0.05-g biomass was added into a 100-mL conical flask with rubber stopper containing 50 mL dye solution, and the pH of the solution was kept at 6.0 adjusted by HCl and NaOH addition. In the adsorption isotherm experiments, the initial concentration ranges used were

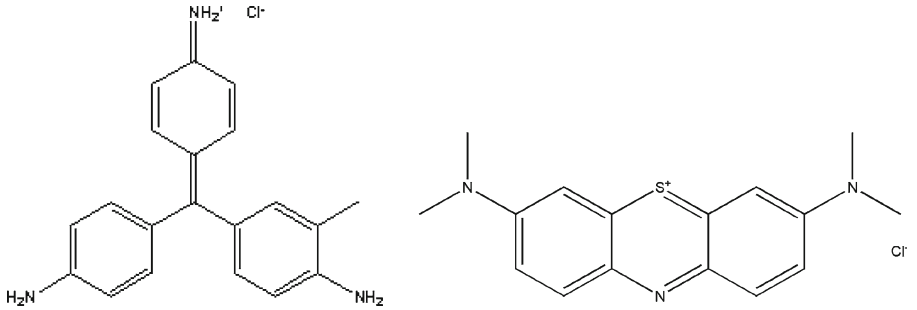

Fig. 2 Structures of methylene blue and basic magenta

0–3,000 mg L^{-1}. In the pH, ionic strength, temperature, and sorption kinetic experiments, the initial concentrations of methylene blue and basic magenta were 380 and 340 mg L^{-1}, respectively. In the ionic strength experiments, different amounts of solid sodium chloride and magnesium chloride were added into the dye solutions to control the ion concentration. After sorption, the biomass was separated from the solution by centrifugation, the concentration of the dyes in the filtrate was determined by measuring their characteristic absorbance by UV–Vis spectrometer (Purkinje General, China), and the characteristic absorbances of MB and BM are 670 and 550 nm, respectively. All the experiments were conducted in duplicate, and the mean values were reported.

Results and Discussion

Characterization of the Biosorbent

Figure 3 shows the TEM image of the modified biomass. It could be seen that the cell wall was kept intact after modification. The cell walls of baker's yeast are mainly composed of mannan, glucans, chitin, and mannoproteins which are abundant sources of different functional groups such as imide, amine, and hydroxyl. XPS was used to characterize the surfaces of the biomass before and after modification. Figure 4 shows typical wide-scan spectra. Three peaks for C 1s, N 1s, and O 1s are observed. The atomic ratios (calculated from the peak area) of C/N/O were 78.3:20.1:1.6 for the unmodified biomass, while those for the modified biomass were 74.0:24.0:2.0. The change in the ratios of oxygen and nitrogen atoms to carbon atom demonstrated the success of the modification.

In order to determine the amounts of the functional groups on the biomass surface, potentiometric titration experiment was carried out. Figure 5 shows the potentiometric titration curve of the modified biomass. It could be seen that there were several inflections in the curve. Since it is difficult to calculate the concentrations of the different functional groups, the first derivative method was used to deal with the titration curve.

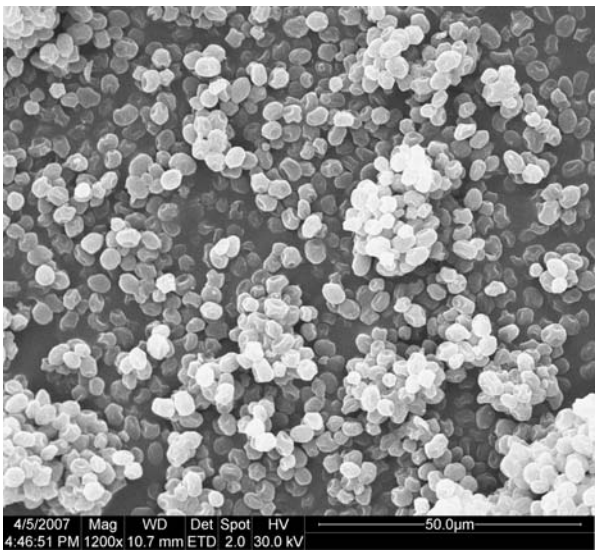

Fig. 3 TEM image of the modified biomass

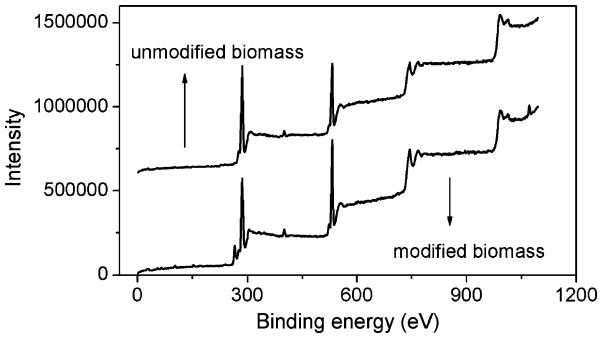

Fig. 4 XPS of the modified and unmodified biomass

First Derivative Method $\left(\Delta\mathrm{pH}/\Delta V - \overline{V}\right)$

In the first derivative method, $\overline{V}$ is the average volume (milliliter), $\Delta\mathrm{pH}/\Delta V$ represents the changes of the pH value with the addition of the titrant, and it could be calculated by the following equation.

$$\frac{\Delta\mathrm{pH}}{\Delta V} = \frac{(\mathrm{pH})_{V_2} - (\mathrm{pH})_{V_1}}{V_2 - V_1} \tag{1}$$

The first derivative method is based on the mathematical principle that an inflection of a curve corresponds to the maximum point of the first derivative curve. The stoichiometric point in the titration curve could be determined through this method. The quantity of the functional groups per gram of biomass could be calculated through the equation below:

$$[\text{functional groups}] = \frac{V_{\mathrm{HCl}} \times C_{\mathrm{HCl}}}{M} \tag{2}$$

where M (gram) is the mass of the modified biomass, V_{HCl} is the volume of the HCl consumed at the stoichiometric point, and C_{HCl} is the concentration of the titrant. The

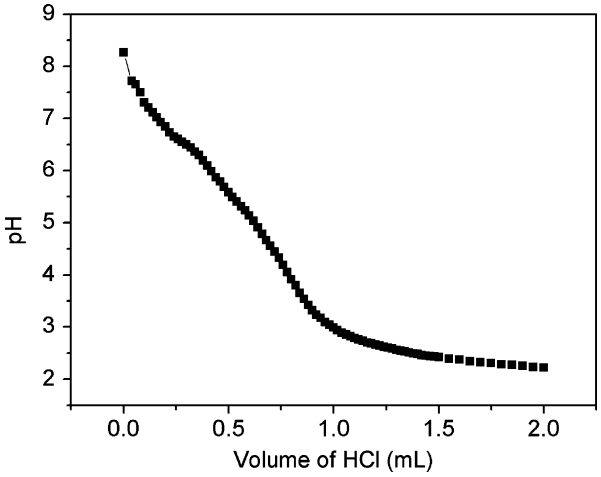

Fig. 5 Potentiometric titration curve of the modified biomass

equilibrium constants (K_a) of the functional groups could also be evaluated through the first derivative curve. K_a is defined as follows:

$$K_a = \frac{[H^+] \cdot [A^-]}{[HA]} \tag{3}$$

where A^- is the functional group, and HA is the protonation form. When the concentration of A^- is equal to that of HA, the value of pK_a is equal to the value of pH. The K_a and the concentration of the functional groups could be determined by the first derivative method.

Figure 6 shows the first derivative curve of the modified biomass. It could be seen that three maximum points were present, demonstrating that there were three different functional groups on the modified biomass surface, which may correspond to amine, imide, and carboxyl groups. The volumes of the titrant (HCl) at the three stoichiometric points were 0.09, 0.45, and 0.81 mL. According to the first derivative method, the concentrations for the three functional groups were 0.27, 1.08, and 1.08 mmol g^{-1}, and the pK_a calculated were 7.7, 6.6, and 5.0, which were similar to that of amine, imide, and carboxyl groups [26].

Effect of pH

pH is one of the parameters that significantly influenced the dye adsorption, and it would affect the interaction between sorbate and sorbent in two ways [27]. Firstly, it would change the condition of the dye ions. Since dyes are complex aromatic organic compounds having different functional groups and unsaturated bonds, they have different ionization potentials at different pH, resulting in the pH-dependent net charge on dye molecules. Secondly, the surface of the biosorbent consists of biopolymers with many functional groups, so the net charge on biosorbent is also pH-dependent. Therefore, the interaction between dye molecules and biosorbent is basically a combined result of charges on dye molecules and the surface of the biosorbent.

pH experiment was conducted at 25 °C. A 0.05-g biomass was added into 50 mL dye solution, and the initial concentrations of methylene blue and basic magenta were 380 and 340 mg L^{-1}, respectively. The pH of the solution was adjusted by HCl and NaOH addition. Figure 7 shows the effect of pH on basic magenta and methylene blue adsorption on the

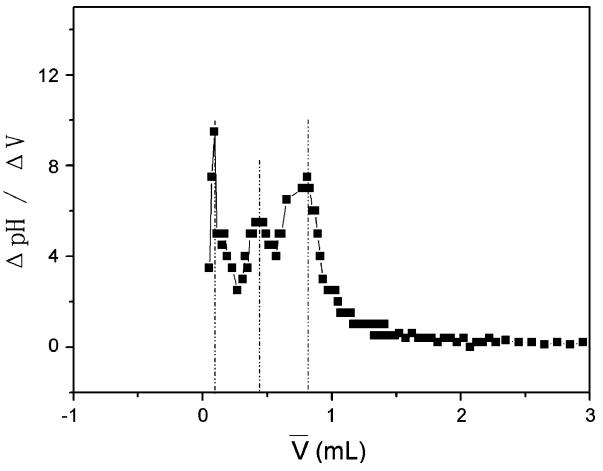

Fig. 6 First derivative curve of the modified biomass

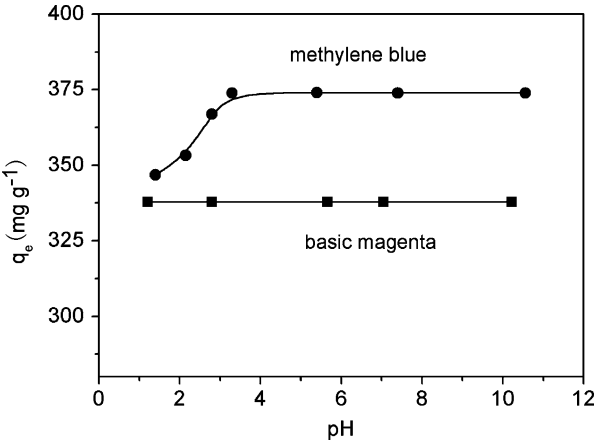

Fig. 7 Effect of pH on the basic magenta and methylene blue adsorption on the modified biomass

modified biomass. It could be seen that pH had little effect on the adsorption of the two dyes over a wide range from 1 to 11. These behaviors of dye sorption could be explained on the basis of the interaction between dye molecules and biosorbent. We know that there were great amounts of functional groups on the surface of the biomass after modification, and they would have much net negative charge in the aqueous phase. The interaction between the negatively charged biosorbent and the two cationic dyes was mainly through attractive electrostatic force. At low solution pH, the net negative charge of the biosorbent would decrease, but the net positive charge on the dye molecule increased because of the protonation amine groups. At high solution pH, although the net electropositivity of the dye molecule decreased, the net electronegativity of the biosorbent would increase due to deprotonation of the functional groups. The interaction between the biosorbent and the dyes would not affect greatly both cases discussed above. The modified biomass kept high adsorption capacity over the pH range from 1 to 11. Literature had reported that the capacity of the unmodified biosorbent for dye reduced greatly with the decrease of the solution pH [28–30], and the advantage of the modified biomass was that it could be used in a wide pH range.

Adsorption Kinetic Experiments

The adsorption kinetic experiments were conducted at the same conditions as in the pH experiment, except that the solution pH is kept at 6.0. The study of adsorption kinetics describes the solute uptake rate. This rate controls the residence time of adsorbate uptake at the solid solution interface. Figure 8 illustrates the adsorption kinetics of the two dyes on the modified biomass. The rates were both rapid during the initial stages of the sorption process. After a very rapid sorption, uptake rates slowly declined with lapse of time and reached equilibrium values at about 40 min for both dyes. Compared to most of biosorbent and porous adsorbents such as activated carbon and resin, the adsorption kinetics of the modified biomass was faster [31]. Hence, a practical advantage of using the modified biomass as an adsorbent would be in its ability to remove more dyes in a much shorter adsorption time. The kinetics of methylene blue and basic magenta biosorption on the modified biomass were analyzed using pseudo-second-order kinetic model.

$$\frac{\mathrm{d}q_t}{\mathrm{d}t} = k_2(q_\mathrm{e} - q_t)^2 \tag{4}$$

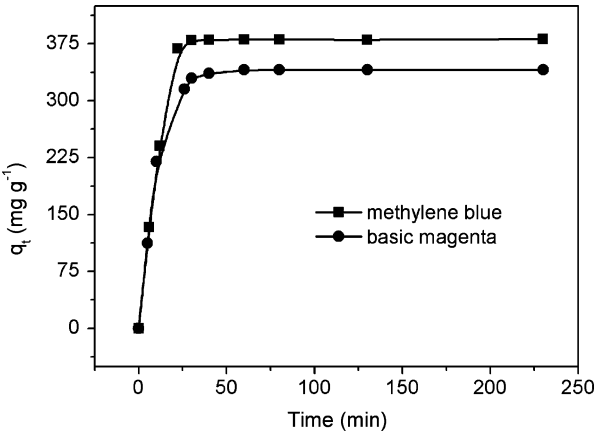

Fig. 8 Adsorption kinetics of basic magenta and methylene blue on the modified biomass

where q_t and q_e are the grams of solute sorbed per gram of sorbent at any time and at equilibrium (milligram per gram), respectively, and k_2 is the rate constant of first-order sorption (gram per milligram per minute). Integrating Eq. 4 for the boundary conditions q_t=0 at t=0 and $q_t = q_t$ at $t = t$ gives:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} = \frac{1}{v_0} + \frac{t}{q_e} \tag{5}$$

Kinetic constants of the model are given in Table 1. It could be seen that the good fit (R^2>0.99) were obtained for both dyes, which indicated that the adsorption conformed to the pseudo-second-order reaction mechanism, illustrating that there were two factors, the amount of active sites and the concentration of the dyes, that control the adsorption rate. Surface modification could increase the amount of the active sites, which would improve the sorption rate.

Adsorption Isotherm Experiments

Studies on the adsorption isotherm are a prerequisite to understand the adsorbate–adsorbent interaction and to optimize the use of the adsorbent. Figure 9 shows the adsorption isotherms of basic magenta and methylene blue on the modified and unmodified biomass. It was observed that the amount of the adsorbed dyes increased with increase in equilibrium concentration and ultimately attained a saturated value. The adsorption data for the modified and unmodified biomass were analyzed using Langmuir and Freundlich adsorption isotherms. The linear Langmuir and Freundlich equations were shown as follows:

$$\frac{C_e}{q_e} = \frac{1}{b \times q_m} + \frac{C_e}{q_m} \tag{6}$$

Table 1 Kinetic parameters for basic magenta and methylene blue adsorption on the modified biomass.

	v_0 (g mg^{-1} min^{-1})	q_e (mg g^{-1})	R^2
Methylene blue	15.7	374.5	0.997
Basic magenta	3.40	327.8	0.996

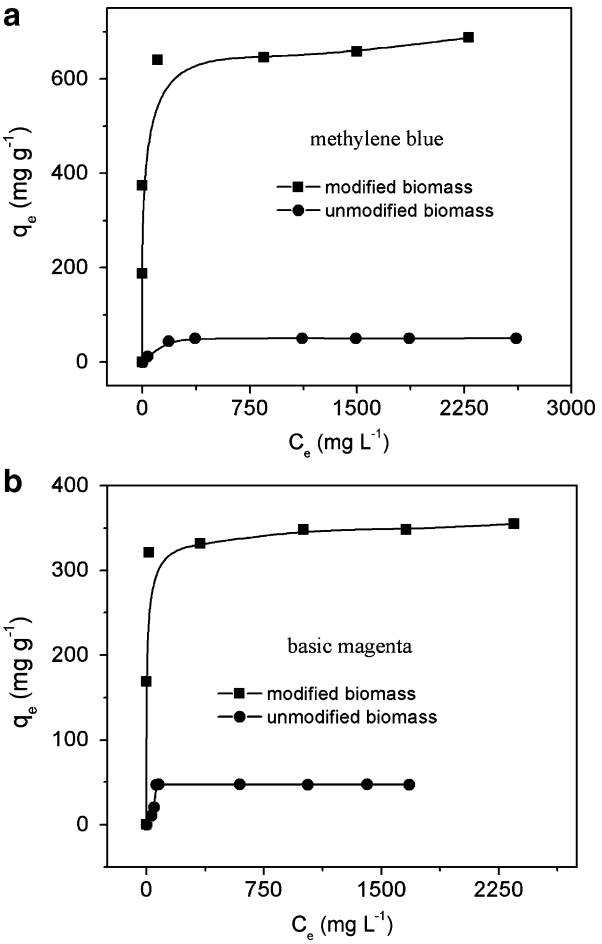

Fig. 9 Adsorption isotherms of basic magenta (**b**) and methylene blue (**a**) on the modified biomass

where q_m is the maximum amount of adsorption (milligram per gram), b is the adsorption equilibrium constant (liter per milligram), and C_e is the equilibrium concentration of substrates in the solution (milligram per liter).

$$\ln q_e = \ln a + \frac{1}{n} \ln C_e \tag{7}$$

where a (in mg$^{1-1/n}$ L$^{1/n}$ g^{-1}) is a constant representing the adsorption capacity, and n is a constant depicting the adsorption intensity. The Langmuir and Freundlich adsorption constants evaluated from the isotherms with the correlation coefficients are listed in Table 2. It can be seen that the Langmuir model gave better fits than the Freundlich model, illustrating that the adsorption on the modified and unmodified biomass was monolayer adsorption. According to the Langmuir equation, the maximum uptake capacities (q_m) of the unmodified biomass for methylene blue and basic magenta were 51.5 and 48.7 mg g^{-1}, while that of the modified biomass were 680.3 and 353.4 mg g^{-1}, respectively. The adsorption capacities of the biomass for methylene blue and basic magenta increased about 13 and seven times after modification.

Table 2 The constants of Langmuir and Freundlich isotherms for basic magenta and methylene blue adsorption.

Sorbents	Dyes	Langmuir			Freundlich		
		q_m (mg g^{-1})	b (L mg^{-1})	R^2	a ($mg^{1-1/n}$ $L^{1/n}$ g^{-1})	n	R^2
Unmodified biomass	Methylene blue	51.5	0.02	0.999	1.3	3.4	0.828
	Basic magenta	48.7	0.02	0.999	1.2	4.4	0.653
Modified biomass	Methylene blue	680.3	0.07	0.999	1.1	10.4	0.910
	Basic magenta	353.4	0.09	0.998	1.1	14.1	0.933

The adsorption of cationic dyes on the biosorbents was mainly attributed to electrostatic interaction and other weak physical forces such as hydrogen bonding and van der Waals interactions. After modification of the biomass with poly(amic acid), large amounts of functional groups such as amine, imide, and carboxyl groups were introduced, which provided more active sites and made the adsorption capacity increased significantly.

Effect of Temperature on the Adsorption

Figure 10 shows the effect of temperature on the adsorption capacity of the modified biomass for the two dyes. It could be seen that the uptakes of the two dyes both increased with increasing temperature up to 35 °C showing the endothermic character of biosorption. Temperature could influence the adsorption step and consequently the reversibility of the desorption equilibrium. In general, an increase in temperature is followed by an increase in the diffusivity of the ion and, consequently, by an increase in the adsorption rate if diffusion is the rate controlling step. The increase in biosorption could be due to increased surface activity and increased kinetic energy of the dye molecules.

Effect of Ionic Strength on the Adsorption

Wastewater from textile-manufacturing or dye-producing industries contains various types of suspended and dissolved compounds apart from the dyes which can be considered as

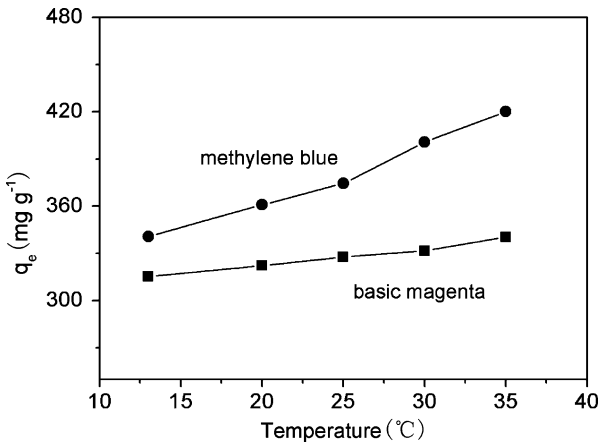

Fig. 10 Effect of temperature on dyes adsorption on the modified biomass

impurities in the dye removal process. Cations such as Na^+, K^+, Mg^{2+}, and Ca^{2+} are the most common metal ions present in wastewater. Literature had reported that the presence of these ions leads to high ionic strength, which may significantly affect the performance of the biosorption process. The effect of ionic strength on the uptake of basic magenta and methylene blue on the modified biomass was also studied in the presence of different ions at different concentrations. Figure 11 presents that ionic strength had little effect on the adsorption of the two dyes at the investigated concentration. As we have known that there are great amounts of functional groups such as carboxyl and amine groups on the biomass after modification, the interaction between the dye molecule and the biomass became very complex. One molecule could adsorb on the biomass surface through several different interactions such as hydrogen bond and attractive electrostatic force at the same time, and the modified biomass had high affinity for the dyes.

Conclusion

Poly(amic acid)-modified biomass was prepared through a simple method. The isotherm experiment showed that the adsorption capacity of the modified biomass for methylene blue

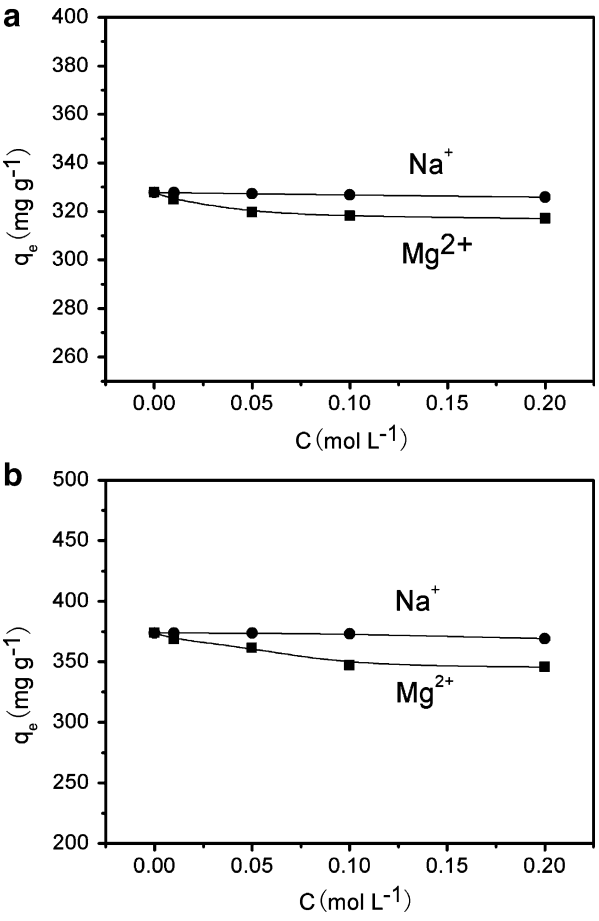

Fig. 11 Effects of ion strength on the adsorption of (**a**) basic magenta and (**b**) methylene blue on the modified biomass

and basic magenta showed a significant increase compared with the pristine biomass due to the presence of a large number of carboxyl groups. The kinetics experiment illustrated that the adsorption process can be completed in a much shorter time. pH and ionic strength had comparatively little effect on the adsorption. The obtained biomass has great potential for practical use.

References

1. Garg, V. K., Amita, M., Kumar, R., & Gupta, R. (2004). *Dyes and Pigments*, *63*, 243–150. doi:10.1016/j.dyepig.2004.03.005.
2. Pearce, C. I., Lloyd, J. R., & Guthrie, J. T. (2003). *Dyes and Pigments*, *58*, 179–196. doi:10.1016/S0143-7208(03)00064-0.
3. Sivaraj, R., Namasivayam, C., & Kadirvelu, K. (2001). *Waste Management (New York, N.Y.)*, *21*(1), 105–110. doi:10.1016/S0956-053X(00)00076-3.
4. Nacera, Y., & Aicha, B. (2006). *Chemical Engineering Journal*, *119*, 121–125. doi:10.1016/j.cej.2006.01.018.
5. Bhatnagar, A., Jain, A. K., & Mukul, M. K. (2005). *Environmental Chemistry Letters*, *2*, 199–202. doi:10.1007/s10311-004-0097-0.
6. Banat, I. M., Nigam, P., Singgh, D., & Marchant, R. (1996). *Bioresource Technology*, *58*(3), 217–227. doi:10.1016/S0960-8524(96)00113-7.
7. Maurya, N. S., Mittal, A. K., Cornel, P., & Rother, E. (2006). *Bioresource Technology*, *97*, 512–521. doi:10.1016/j.biortech.2005.02.045.
8. Flavio, A. P., Yoshitaka, G., Ana, C. M., Silvio, L. P. D., & Eder, C. L. (2007). *Dyes and Pigments*, *72*, 256–266. doi:10.1016/j.dyepig.2005.09.001.
9. Han, R., Wang, Y., Han, P., Shi, J., Yang, J., & Lu, Y. (2006). *Journal of Hazardous Materials*, *137*, 550–557. doi:10.1016/j.jhazmat.2006.02.029.
10. Forgacs, E., Csehati, T., & Oros, G. (2004). *Environment International*, *30*, 953–971. doi:10.1016/j.envint.2004.02.001.
11. Aksu, Z. (2005). *Process Biochemistry*, *40*, 997–1026. doi:10.1016/j.procbio.2004.04.008.
12. Aksu, Z., & Tezer, S. (2005). *Process Biochemistry*, *40*, 1347–1361. doi:10.1016/j.procbio.2004.06.007.
13. Tan, T. W., & Cheng, P. (2003). *Applied Biochemistry and Biotechnology*, *104*, 119–128. doi:10.1385/ABAB:104:2:119.
14. Gulati, R., Saxena, R. K., & Gupta, R. (2002). *World Journal of Microbiology & Biotechnology*, *18*, 397–401. doi:10.1023/A:1015540921432.
15. Das, S. K., Bhowal, J., Das, A. R., & Guha, A. K. (2006). *Langmuir*, *22*, 7265–7272. doi:10.1021/la0526378.
16. Deng, S. B., & Ting, Y. P. (2005). *Langmuir*, *21*, 5940–5948. doi:10.1021/la047349a.
17. Chen, J. P., & Yang, L. (2005). *Industrial & Engineering Chemistry Research*, *44*, 9931–9942. doi:10.1021/ie050678t.
18. Deng, S. B., & Ting, Y. P. (2005). *Water Research*, *39*, 2167–2177. doi:10.1016/j.watres.2005.03.033.
19. Matis, K. A., Loukidou, M. X., Zouboulis, A. I., & Maria, L. K. (2003). *Water Research*, *37*, 4544–4552. doi:10.1016/S0043-1354(03)00415-9.
20. Rad, R. J., Ghafourian, H., Asef, Y., Dalir, S. T., Sahafipour, M. H., & Gharanjik, B. M. (2004). *Journal of Hazardous Materials*, *116*, 125–134. doi:10.1016/j.jhazmat.2004.08.022.
21. Yu, J. X., Tong, M., Sun, X. M., & Li, B. H. (2007). *Journal of Hazardous Materials*, *143*, 277–284. doi:10.1016/j.jhazmat.2006.09.021.
22. Yu, J. X., Tong, M., Sun, X. M., & Li, B. H. (2007). *Biochemical Engineering Journal*, *33*, 126–133. doi:10.1016/j.bej.2006.10.012.
23. Reddad, Z., Gerente, C., Andres, Y., & Le Cloirec, P. (2002). *Environmental Science & Technology*, *36* (10), 2242–2248. doi:10.1021/es010237a.
24. Pagnanelli, F., Papini, M. P., Toro, L., Trifoni, M., & Veglio, F. (2000). *Environmental Science & Technology*, *34*(13), 2773–2778. doi:10.1021/es991271g.
25. Yee, N., Benning, L. G., Phoenix, V. R., & Ferris, F. G. (2004). *Environmental Science & Technology*, *38* (3), 775–782. doi:10.1021/es0346680.
26. Wuhan University (2000). *Analytical chemistry* (pp. 319–320, 4th ed.). Wuhan, China: Higher Education Press.
27. Maurya, N. S., Mittal, A. K., Cornel, P., & Rother, E. (2006). *Bioresource Technology*, *97*, 512–521. doi:10.1016/j.biortech.2005.02.045.

28. Ozer, A., Akkaya, G., & Turabik, M. (2006). *Journal of Hazardous Materials*, *135*, 355–364. doi:10.1016/j.jhazmat.2005.11.080.
29. Aksu, Z. (2006). *Separation and Purification Technology*, *48*, 24–35. doi:10.1016/j.seppur.2005.07.017.
30. Won, S. W., Kim, H. J., Choi, S. H., Chung, B. W., Kim, K. J., & Yun, Y. S. (2006). *Chemical Engineering Journal*, *121*, 37–43. doi:10.1016/j.cej.2006.04.005.
31. Eigeundi, M. S. (1991). *Water Research*, *25*(3), 271–273. doi:10.1016/0043-1354(91)90006-C.