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Algae removal from raw water by flocculation and the fractal characteristics of flocs

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ABSTRACT

The feasibility of flocculation treatment of algae-containing raw water using poly(acrylamide-acryloyloxyethyl trimethyl ammonium chloride-butyl acrylate) (PADB) was investigated. The effects of flocculant dosage and initial pH of raw wastewater on the removal of turbidity and chlorophyll-a (Chl-a) from raw water were examined. The flocculation efficiency of PADB was compared with that of cationic polyacrylamide (CPAM), polymeric ferric sulfate (PFS), and polyaluminum chloride (PAC). PADBs proved to be highly efficient flocculants for the removal of Chl-a; meanwhile, flocculation tests also demonstrated the superiority of PADB over CPAM, PFS, and PAC in the removal of Chl-a. The removal of turbidity and Chl-a by PADB were higher than 94 and 99%, respectively, in the pH range of 6–8 at 3 mg L^{-1} PABD dosage and at 40% cationic degree. The Chl-a removal efficiency of PADB was 7.6, 17.9, and 52.8% higher than that of CPAM, PFS, and PAC, respectively. Optical microscopy was used to investigate the fractal characteristic of flocs formed during flocculation. The fractal dimension increased, and the flocs increased in size and density with increasing pH. In conclusion, the flocculation-separation process using PADBs as flocculants proved to be highly effective and efficient method for algae removal, and provided guidance and directions for future practical application.

Keywords: Flocculant; Flocculation; Algae; Removal efficiency; Fractal dimension

1. Introduction

Algae-related problems in water treatment, eutrophication in particular, have become a worldwide issue. The occurrence of toxic algae in water bodies used as production sources of drinking water is a global problem [1]. Eutrophication refers to the discharge of large quantities of nitrogen, phosphorus, and other nutrients into reservoirs and lakes, leading to the deterioration of ecological structure and function in the water environment. Eutrophication leads to rapid growth and reproduction of algae, which causes algal blooms. Harmful algal blooms (HABs) pose a serious threat to aquatic life, human health, local tourism, and coastal aesthetics. Moreover, some HABs produce

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toxins. More than half of the fresh waters in China suffer from HABs [2]. As of this writing, more than 60% of the lakes in China, especially Taihu Lake, are eutrophicated, and are suffering from HABs; cyanobacteria *Microcystis aeruginosa* (MA) is one of the dominant species [3]. Meanwhile, HABs have also become common along the coastline of California for many years [4]. The excessive growth of toxic MA greatly reduced water quality, damaged the natural functions of lakes, and threatened the drinking water resources and the city's safe water supply. HABs are occurring with greater frequency and intensity in many shallow lakes. Severe HAB occurred in 700 km^2 of Taihu Lake (China) in 2007, giving rise to a drinking water crisis in Wuxi City [5]. Developing HAB control techniques that are safe, highly efficient, and cost effective is among the top priorities in China [6].

In the past years, numerous technological approaches around the world were developed to solve the HAB problem. Such approaches include floatation, filtration, and pumping/sucking, algaecides, filter-feeding fish, zoon plankton control, plant allelopathy or bacteria, and flocculation [7–9]. Treatment approaches can remove HABs temporarily, but a variety of adverse effects may arise, including change of pH and oxygen levels, and adverse ecological effects; these technologies also require high flocculant dosage and high costs, and may produce a large volume of residual sludge [5].

Most microalgae removal methods involve economical and technical drawbacks, but the flocculation–coagulation process is convenient because large quantities of cultures can be treated [10]. Traditional flocculants, such as metal and mineral salts, and chitosan, have been used for algae removal [7,11]. Flocculation by metal salts, namely $FeCl_3$ and $Al_2(SO_4)_3$ may be unacceptable because of low removal efficiency, and the supernatant produced after flocculation may cause discoloration. The flocculation efficiencies of $FeCl_3$ and $Al_2(SO_4)_3$ were 53.31 and 49.74% at rates of 150 and 100 $mg\,L^{-1}$, respectively, under neutral conditions [12]. A major limitation of using mineral salts, such as montmorillonite, kaolinite, and phosphatic salt, is that high flocculant dosages are required, ranging from 120 to 1,000 $mg\,L^{-1}$; a large volume of sludge production and subsequent processing fee may also be involved in the processes [13]. Microalgae cells are generally negatively charged; thus, the use of cationic polymers is recommended for coagulation–flocculation [14]. Most studies conducted as of this writing assessed the potential of conventional aluminum or ferric salts for microalgae removal, but information is lacking on the new generation of high-performance polymeric flocculants [15].

In this paper, we evaluated the ability of cationic flocculants, including poly (acrylamide—acryloyloxyethyl trimethyl ammonium chloride—butyl acrylate) (PADB), to remove algae from raw water. Turbidity and chlorophyll-a (Chl-a) removal were used as indexes to evaluate the performance of flocculants. The effects of three commercial polymeric flocculants, namely cationic polyacrylamide (CPAM), polymeric ferric sulfate (PFS), and polyaluminum chloride (PAC), were compared with those of PADB. The fractal dimensions of PADB under different conditions were investigated. In addition, the effects of the flocculant dosage and pH values of raw water on the algae removal efficiencies and fractal dimensions of these compounds were discussed.

2. Experimental

2.1. Material

PADB, a CPAM, was synthesized in the laboratory. The detailed synthesis and characterization of PADB were previously reported [16,17]. CPAM with a cationic degree (CD) of 35% and molecular weight (MW) of 525×10^4 was obtained from HaiXia Chemical Co., Ltd (Zhengjiang, China). PAFS and PAC were sourced from Chongqing LanJie Tap Water Material Co., Ltd (Chongqing, China). All reagents used in this study were of analytical grade and were used without further purification. The aqueous and standard solutions were prepared with deionized water. The characteristics of PADB (with different CDs) and CPAM are shown in Table 1.

2.2. Water sample

Raw water containing algae was obtained from Minzhu Lake in Chongqing University (Chongqing, China). The raw water was light green in color, highly turbid, and contaminated with suspended solids. Raw wastewater was filtered through a graticule mesh to remove all large particles and phytoplankton. The flocculation efficiency was determined by measuring

Table 1
Characteristics of organic flocculants used in the experiments

Flocculant	CD (%)	MW (10^4)
PADB1	20	500
PADB2	40	500
CPAM	35	525

turbidity and Chl-a content. Water chemical analysis showed that the turbidity, Chl-a content, and pH values were 30.1–37.5 NTU, 18.0–78.0 μg L^{-1}, and pH 7.0–8.0, respectively.

2.3. Flocculation tests

In this study, turbidity was selected as an index to evaluate the flocculation efficiency, whereas the Chl-a content was used as an index to investigate the removal of algae in the raw water by flocculation. All flocculation experiments were conducted in 1.0 L Plexiglass beakers using a program-controlled jar-test apparatus (ZR4-6, Zhongrun Water Industry Technology Development Co., Ltd, China) at room temperature. The pH of raw wastewater was adjusted with HCl (0.1 mol L^{-1}) or NaOH (0.1 mol L^{-1}). During the rapid stirring phase, a measured amount of flocculant was pipetted into the raw water sample (1.0 L). The dosages of different flocculants were calculated by the quantity of their effective component, i.e. PFS by Fe, PAC by Al, and PADB and CPAM by dry weight. The raw water samples were mixed rapidly at 120 rpm for 3 min after treatment, followed by slow stirring at 40 rpm for 6 min, and sedimentation for 10 min. After sedimentation, the supernatant samples were collected at 2 cm below the surface of the tested water sample to measure turbidity (2100Q Turbidimeter, HACH, USA) and Chl-a (TU-901 Double Beam UV–visible spectrophotometer, Beijing General Instrument Co., Ltd, China). The tests were conducted three times, and the relative error was less than 5%. Meanwhile, the obtained green algae flocs were observed under the microscope (Metallographic Microscope MIT300, Chongqing OTT Optical Instrument Co., Ltd, China).

2.4. Analytical methods

2.4.1. Determination of Chl-a

Chlorophyll-a method was the common way to determine the algae content and evaluate algae-removing efficiency. The Chlorophyll-a used as an index to investigate the removal of algae has been found to be significant and common according to those reported in the previous literature [2]. Chl-a content was determined according to Chinese national standards and based on Water Quality Determination of Chlorophyll by Spectrophotometry (No. SL88-2012). The test solution at 20 mL was filtered through a 0.45 μm microporous membrane to remove residual plant plankton. The membrane was dissolved completely in 8 mL of 90% acetone solution, which was subsequently centrifuged at a speed of 3,500 rpm for 15 min. The total extraction time was 9.5 h according to Chinese national standard (No. SL88-2012). The supernatant was collected to determine the absorbance at 750, 664, 647, and 630 nm. The concentration of Chl-a ($\rho_{\text{chl-a}}$) and the removal of $\rho_{\text{chl-a}}$ were calculated by the following equations:

$$\rho_{\text{chl-a}} = \frac{[11.85(A_{664} - A_{750}) - 1.54(A_{647} - A_{750}) - 0.08(A_{630} - A_{750})]V_1}{V_2 L} \quad (1)$$

$$\text{Removal}\,(\rho_{\text{chl-a}}) = \frac{\rho_{\text{chl-a}}(\text{before}) - \rho_{\text{chl-a}}(\text{after})}{\rho_{\text{chl-a}}(\text{before})} \times 100\% \quad (2)$$

where $\rho_{\text{chl-a}}$ is the concentration of Chl-a in μg L^{-1}; A_{750} is the absorbance of the extracting solution at 750 nm; A_{664} is the absorbance of the extracting solution at 664 nm; A_{647} is the absorbance of the extracting solution at 647 nm; A_{630} is the absorbance of the extracting solution at 630 nm; V_1 is the volume of the extracting solution in mL; V_2 is the volume of the water sample in L; L is the optical distance of the cuvette in cm; $\rho_{\text{chl-a}}$(before) is the concentration of Chl-a before flocculation in μg L^{-1}; and $\rho_{\text{chl-a}}$(after) is the concentration of Chl-a after flocculation in μg L^{-1}.

2.4.2. Measurement of the fractal dimension

Floc samples were collected after flocculation with PADB under optimum conditions. Floc samples were dried by natural evaporation on a clear ground slide. To study the floc characteristics, the fractal dimensions of flocs were measured by image analysis. Heterogeneously (non-uniformly) packed objects with irregular boundaries can be defined by non-linear relationships in which the properties of the object scale with a characteristic length dimension were raised to a power called the fractal dimension [18]. The fractal dimension is defined by a power law relationship between a projected area (A) and the characteristic length (L) of the aggregates. The fractal dimension can be calculated from reference:

The relationship between projected area and projection perimeter is given by the following formula:

$$A \sim L^D \quad (3)$$

where A is the projected area (μm^2); L is the projection perimeter (μm); and D is the fractal dimension in 2D space. Performing the natural logarithm in Eq. (3), the following form is obtained:

$$\mathrm{Ln}(A) = D\mathrm{Ln}(L) + \mathrm{Ln}(\alpha) \quad (4)$$

In Eq. (4), Ln(A) is a dependent variable and Ln(L) is an independent variable. Plotting Ln(A) against Ln(L) yields a straight line with slope D; and Ln(α) is the Ln (A) intercept. These parameters were measured using the software Image-Pro Plus and a microscope.

3. Results and discussion

3.1. Effect of dosage on the flocculation efficiency

Fig. 1 indicates that the cationic flocculant PADB has a very good purification effect. The effect of dosage on removal of turbidity is shown in Fig. 2. Before the flocculants were added, the raw water was slightly green, and the algae were uniformly dispersed in the raw water. With increasing PADB dosage, the turbidity removal efficiency increased and then slightly decreased. PADB1 and PADB2 at $3.0\,\mathrm{mg\,L^{-1}}$ resulted in optimal turbidity removal efficiencies of 94.1 and 96.4%, respectively. The effect of flocculant dosage on Chl-a removal is shown in Fig. 3. The removal of Chl-a increased with increasing dosage. At a dosage of more than $2.5\,\mathrm{mg\,L^{-1}}$, the removal efficiency of Chl-a increased gradually. The optimal removal efficiencies of Chl-a were 96.2 and 99.5% with PADB1 and PADB2 at $3.0\,\mathrm{mg\,L^{-1}}$, respectively. At PADB dosage of more than $3.0\,\mathrm{mg\,L^{-1}}$, the removal of turbidity and Chl-a slowly decreased.

The above-mentioned phenomena can be explained by various reasons. For instance, the amount of polymer was insufficient and failed to adsorb and bridge

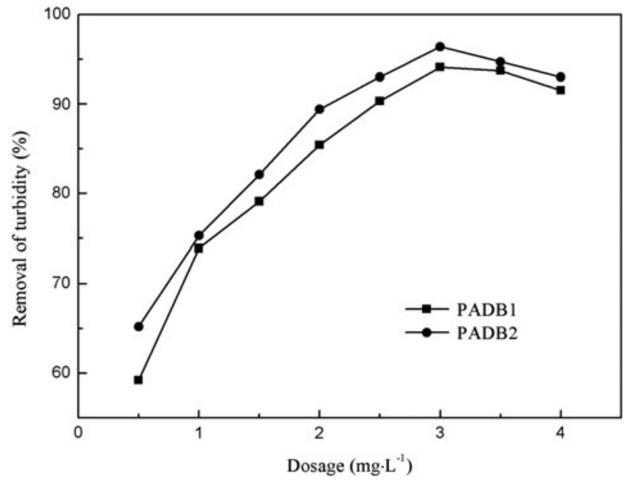

Fig. 2. Effect of PADB dosage on turbidity removal.

the particles to form flocs. With increasing dosage, flocculants were neutralized, or inverted electrical repulsions occurred between microalgal cells, forming large and dense flocs (cell aggregates) by creating bridges between the neutralized microalgae. Thus, the optimal removal efficiency was obtained. At extremely high PADB dosage, the negatively charged algal cells were absorbed by the excess PADB, which results in the negatively charged original algal cells becoming positively charged. Restabilization phenomenon occurred, resulting in the formation of a large number of small and stable flocs that are difficult to precipitate. This phenomenon largely reduced the effectiveness of adsorption–bridging and sweeping–netting effects, negatively affecting the flocculation efficiency.

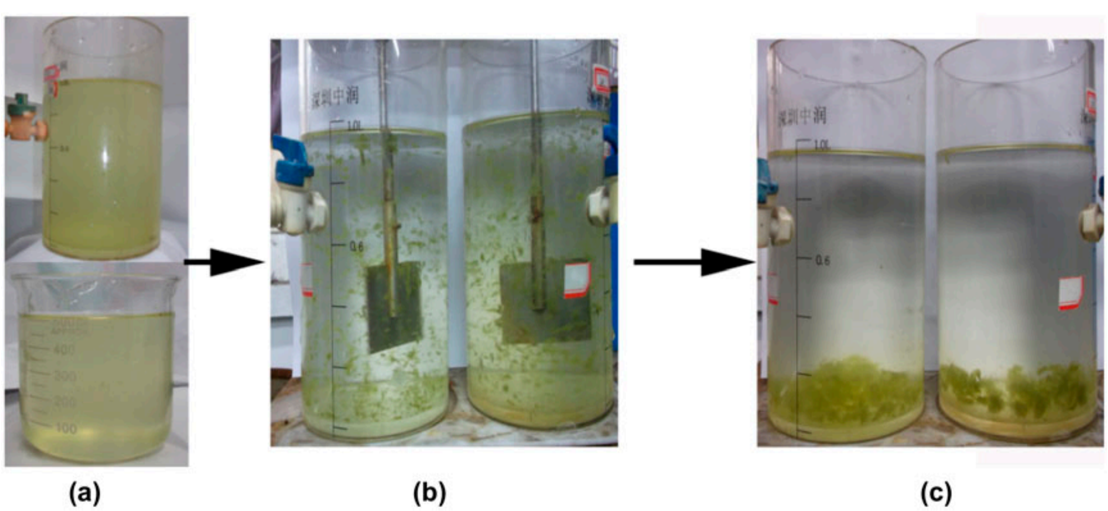

Fig. 1. Flocculation–sedimentation of algal suspension by PADB as follows: (a) raw water, (b) flocculation process, and (c) after sedimentation.

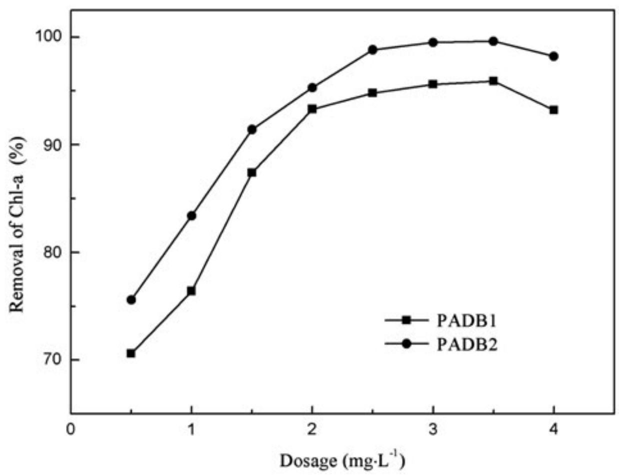

Fig. 3. Effect of PADB dosage on Chl-a removal.

In the algal removal process, charge neutralization and bridging abilities were the main flocculation mechanisms of PADB; PADB had high positive charge density and a long molecular chain [19]. High-CD PADB had high flocculation efficiency. Thus, raw water algicidal removal efficiency can be improved by increasing the CD of PADB. The optimal dosage of PADB for the removal of Chl-a was 3.0 mg L^{-1}.

3.2. Effect of pH value on the removal of Chl-a

As shown in Figs. 4 and 5, turbidity and Chl-a removal rates increased quickly with increasing pH and then decreased sharply. The optimal removal efficiencies water was eighth. The optimal removal rates of turbidity were 91.2 and 94.1% for PADB1 and PADB2, respectively, at pH 7. The optimal removal efficiencies of Chl-a were 95.4 and 99.7% for PADB1 and PADB2, respectively, at pH 8.

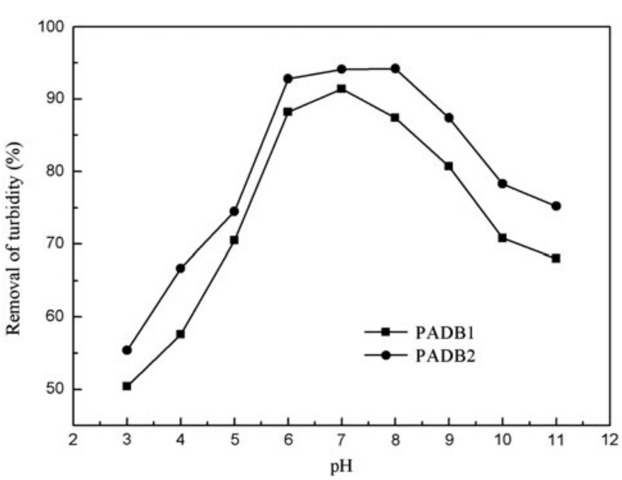

Fig. 4. Effect of pH on removal of turbidity.

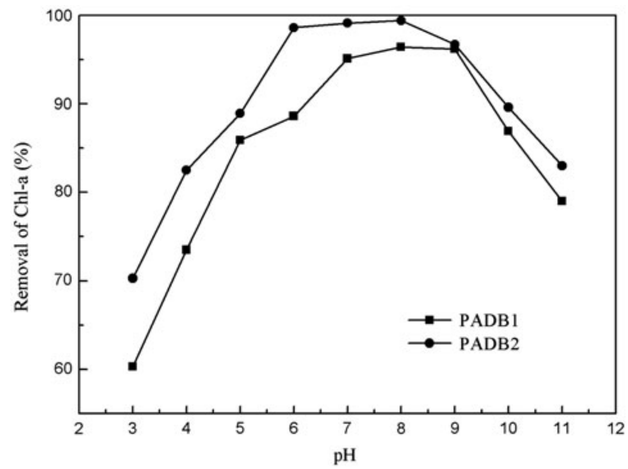

Fig. 5. Effect of pH on removal of Chl-a.

At a low pH value of algae-containing raw water, algae surface charge was reversed and was positively charged by H^+, whereas the PADB was quaternized. Electrostatic repulsion between the algae and cationic groups on the flocculant increased, contributing to the increase in turbidity and Chl-a content [20]. When the pH value was between 6 and 9, the charges of colloidal particles and algae were neutralized, and suspended particles were intensively captured to form adsorption bridges that favor flocculation; flocculation improvement increased the removal efficiencies of turbidity and Chl-a [21]. Under strongly alkaline conditions, the colloidal particles and algae were negatively charged, and the flocculate could not neutralize the negative charge [22]. In addition, the flocculate was hydrolyzed with high pH value, reducing flocculation efficiency. Therefore, pH 6–9 was the optimal range for algae removal.

3.3. Effect of CD on the removal of chlorophyll-a

The effect of CD on turbidity and Chl-a removal is shown in Fig. 6. With increasing CD, a gradual increase in the removal rate of turbidity and Chl-a was observed. The highest removal rates of turbidity and Chl-a were obtained at a CD of 40%. The optimal removal efficiencies of turbidity and Chl-a were 96.2 and 99.8%, respectively. In addition, removal efficiencies of turbidity and Chl-a decreased when CD was higher than 40%.

During development, algae release acidic polysaccharide to the extracellular layer, forming organic

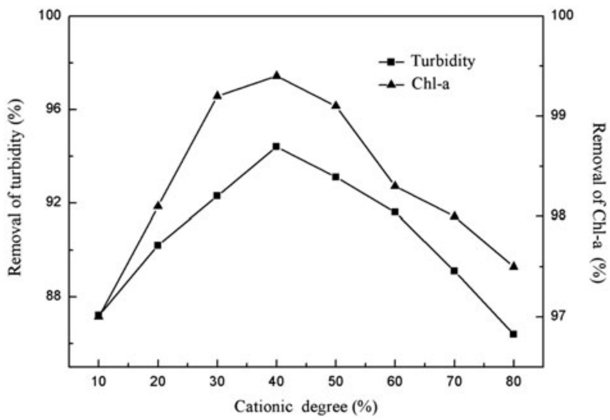

Fig. 6. Effect of CD of PADB on the removal of turbidity and Chl-a.

substances in the extracellular gum and creating a negatively charged algal surface. Thus, algae disperse stably in water by mutual electronic repulsion. At low CD, positive charge was insufficient to destabilize algae and prevent the formation of flocs. At a CD of 40%, electrostatic repulsion increased between polymer chains, resulting in the spread of the polymer chain and improvement of the adsorption–bridging effect [23]. However, a CD higher than 40% was less effective on floc formation because of the presence of charge reversal by the excess positive charge on the algal surface; such charge reversal results in repulsion between the algae and PADB cationic groups [24]. We observed the effects of poor flocculation and sedimentation. Therefore, the best treatment efficiency was achieved at a CD of 40%.

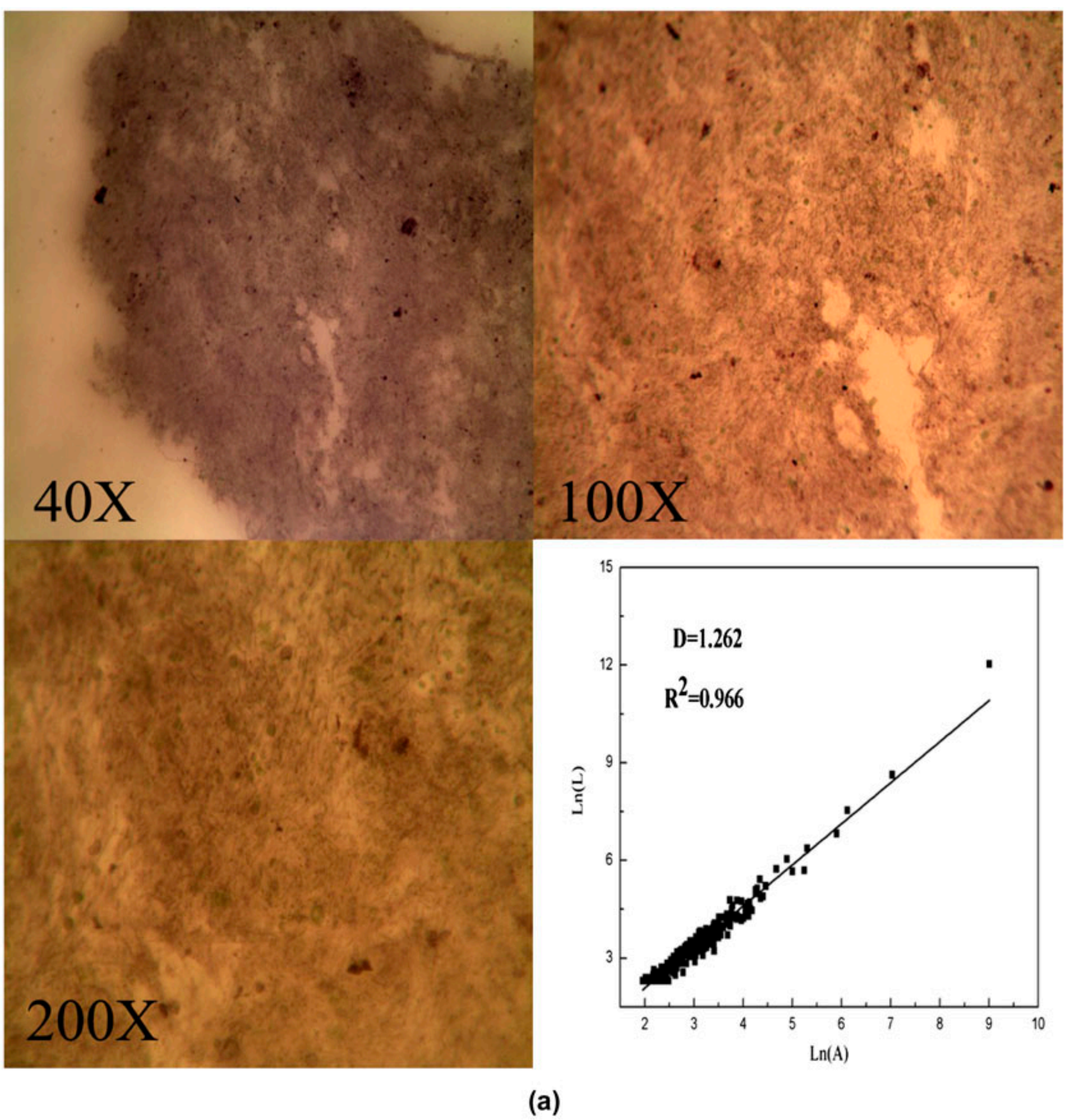

Fig. 7. Microscope image and fractal dimension of flocs flocculated by PADB under the following conditions: (a) pH 3 and 1.5 mg L^{-1}; (b) pH 7 and 1.5 mg L^{-1}; (c) pH 7 and 3.0 mg L^{-1}; and (d) pH 11 and 1.5 mg L^{-1}.

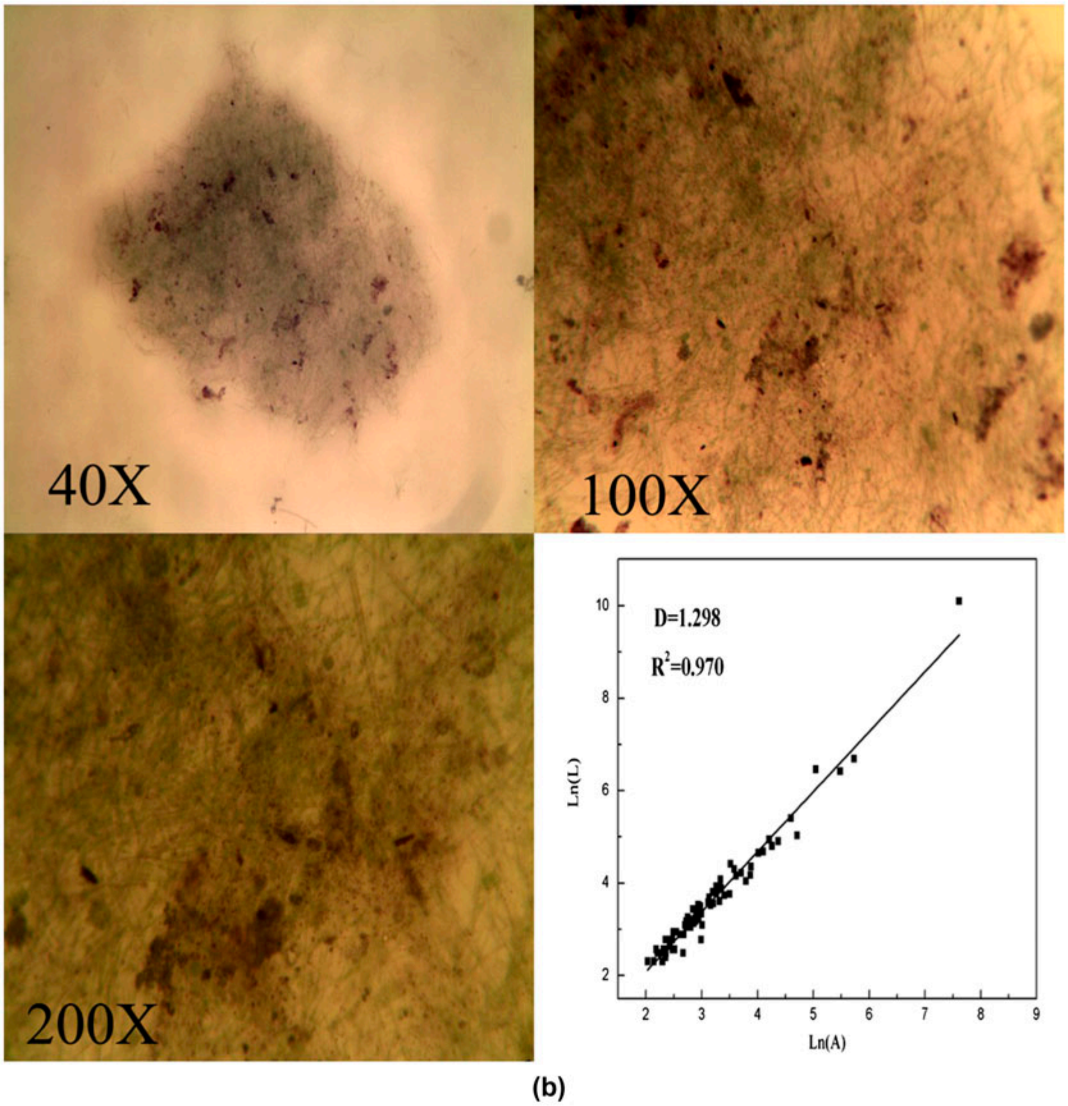

Fig. 7. (*Continued*).

3.4. Fractal dimensions

An alternative way of studying the floc characteristics is macroscopic measurement. According to fractal theory, irregular and porous flocs were geometrically fractal during flocculation. The most important and powerful quantitative parameter is the fractal dimension, which indicates the space-filling capacity, that is the compactness of the floc. A large fractal dimension indicates a compact floc, which is usually preferred in water treatment because low sludge volumes and easy sedimentation are obtained [25]. Fig. 7 shows the microscope image and the fractal dimension of the flocs under the following conditions: (a) pH 3 and 1.5 mg L^{-1}; (b) pH 7 and 1.5 mg L^{-1}; (c) pH 7 and 3.0 mg L^{-1}; and (d) pH 11 and 1.5 mg L^{-1}. Fig. 8 shows the fractal dimension vs. pH value. The algae dispersed in water was flocculated and aggregated in flocs (Fig. 7). The color of algae flocs at pH 3 was light grey and slightly yellow and the algae flocs formed at pH values of 7 and 11 were light green in color. The algae may have been acidized, resulting in their deactivation.

The fractal dimensions of flocs measured under different pH conditions are shown in Fig. 8. The fractal dimension increased with increasing pH value. The fractal dimension was 1.262, 1.298, and 1.358 for pH 3, 7, and 11, respectively, at a dosage of 1.5 mg L^{-1}. The average fractal dimension of the flocs depended on

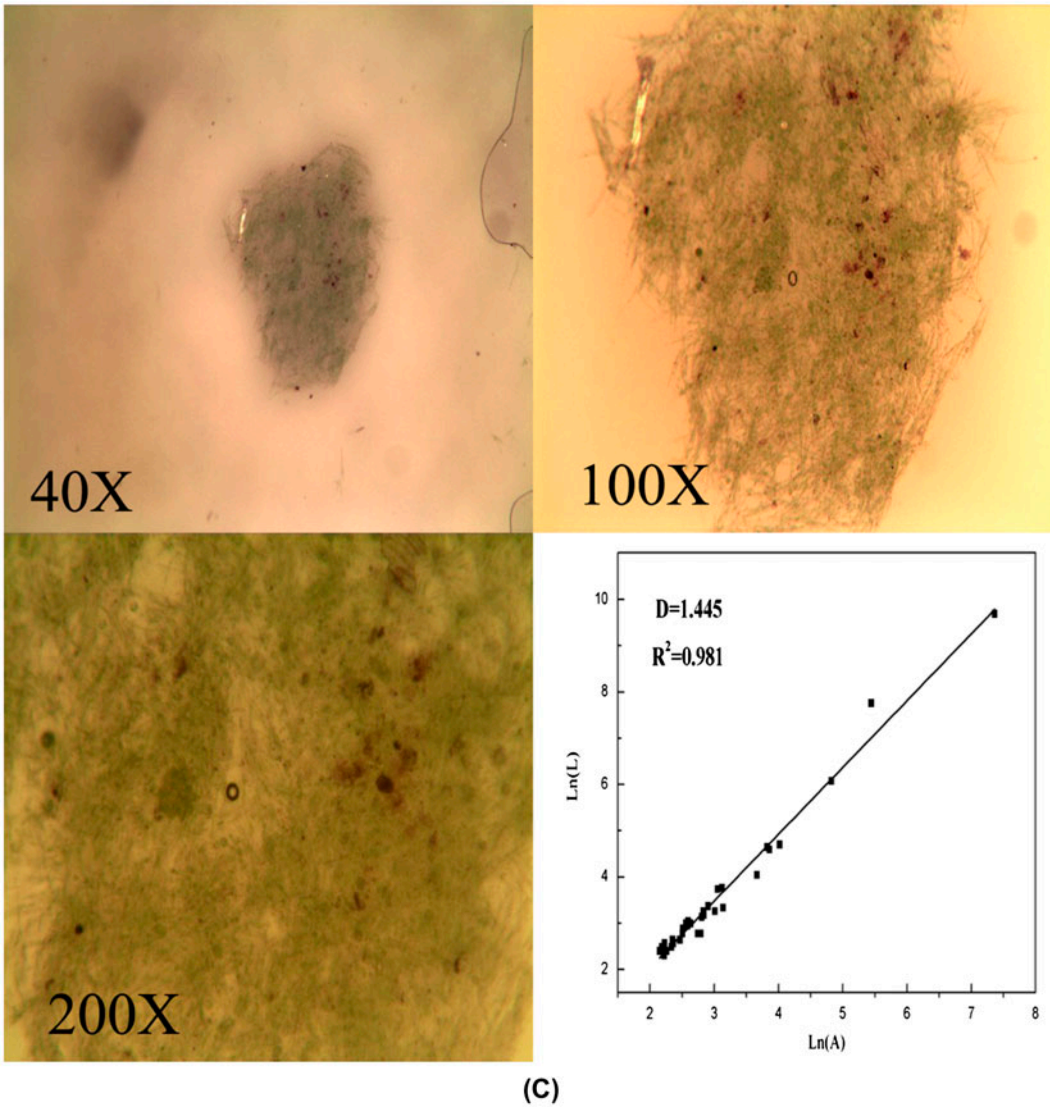

Fig. 7. (*Continued*).

the flocculation conditions and revealed satisfactory self-similarity. Meanwhile, the fractal dimensions at dosages of 1.5 and 3.0 mg L^{-1} were 1.298 and 1.445, respectively. The results of comparison between Fig. 7(b) and (c) indicated that high dosage corresponded to high fractal dimension and large flocs. Given the relationship between fractal dimension and floc characteristics, the flocs with high fractal dimension showed strong density, large cutter size, and high sedimentation rate [26]. Gorczyca and Ganczarczyk [27] reported that the difference in fractal dimensions was due to the various surface charges of the properties of aggregating flocs and the fluid mechanical environment.

3.5. Comparisons between PADB and commercially available flocculants

To compare the flocculation efficiency of different flocculants, some commercially available products, such as CPAM, PFS, and PAC, were used to investigate the algal removal efficiency under different dosages. The results are shown in Figs. 9 and 10. As shown in Figs. 9 and 10, when the dosage was at 3.0 mg L^{-1}, the removal efficiencies of turbidity by PADB1 and CPAM were 94.3 and 92.1%, respectively. The removal efficiencies of turbidity by PFS and PAC were 86.7 and 84.2% at 35 and 40 mg L^{-1}, respectively. The raw water flocculated by PFS was slightly yellow

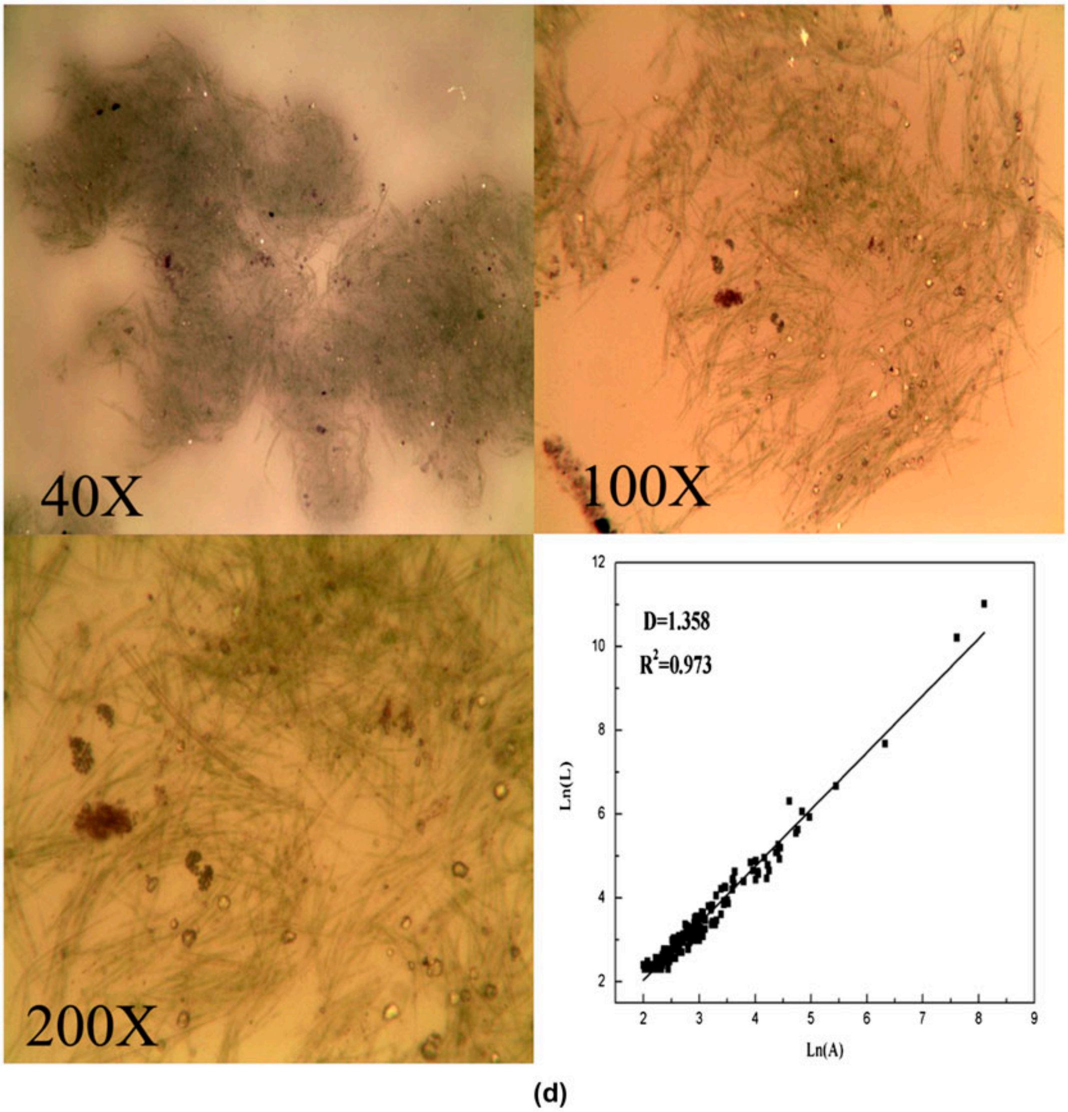

Fig. 7. (*Continued*).

in color because of the residual Fe^{3+} in the water. According to Figs. 9 and 10, the optimal removal of Chl-a by PADB1, CPAM, PFS, and PAC were 99.1, 97.3, 84.6, and 65.3% at dosages of 3.5, 3, 35, and 30 mg L^{-1}, respectively. The removal efficiency of Chl-a by PADB was significantly higher than those by CPAM, PFS, and PAC. In addition, the dosage of PADB and CPAM (about 10% of the dosage of organic flocculant) used in the flocculation process was significantly less than those of PFS and PAC. Meanwhile, after flocculation process, the sludge volume of PADB1, CPAM, PFS, and PAC was 60, 70, 55, and 60 mL. The sludge generated by PADB and CPAM was more loose and porous, while the sludge generated by PFS and PAC was more compact and tighter. However, after adding the mineral salts, including

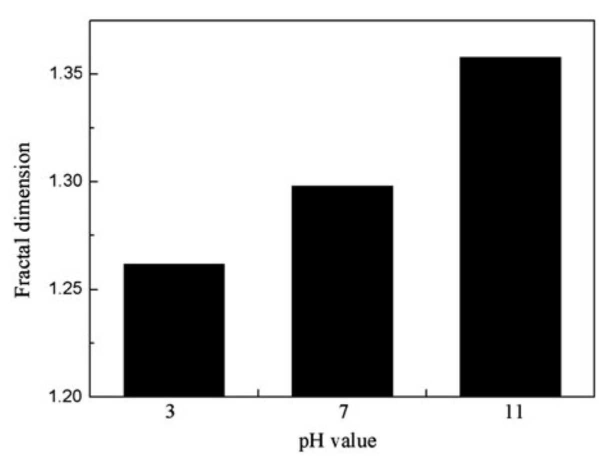

Fig. 8. Fractal dimension vs. pH value flocculated by PADB.

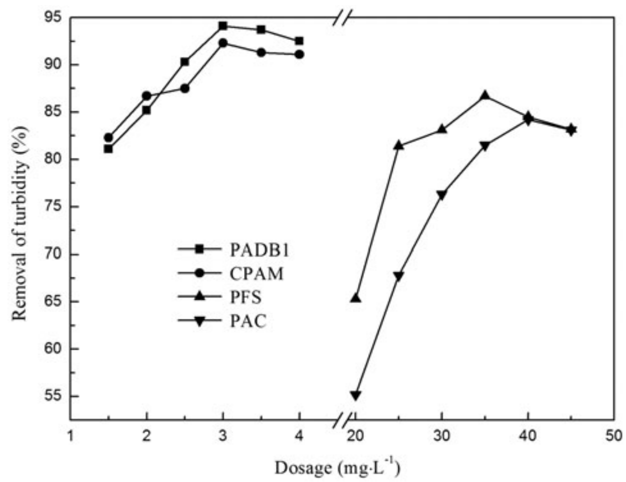

Fig. 9. Comparisons between PADB and commercially available flocculants/coagulants on turbidity removal.

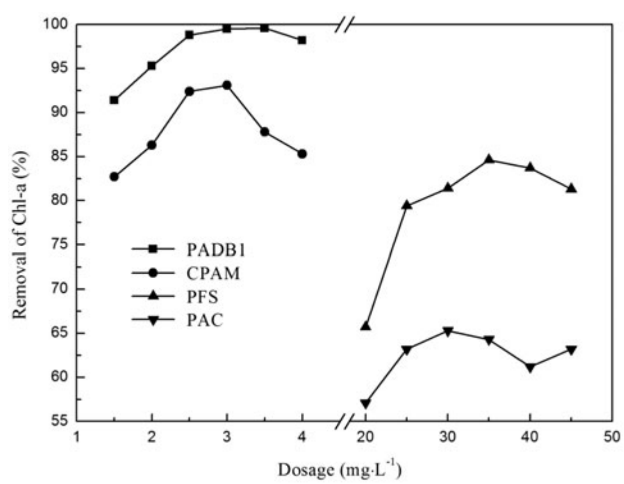

Fig. 10. Comparisons between PADB and commercially available flocculants/coagulants on Chl-a removal.

montmorillonite, kaolinite, and phosphatic salt (monopotassium phosphate), there was no flocculation phenomenon observed. The removal efficiency of the chlorophyll-a (Chl-a) by the mineral salts was about 2–13% at the dosage 100–300 $mg\,L^{-1}$.

4. Conclusions

In this research, the efficiency of turbidity and Chl-a removal by PADB through flocculation was studied. Turbidity and Chl-a were used as indexes to indicate the flocculation efficiency. The following conclusions were obtained:

(1) PADB treatment of algae-containing raw water at 3.0 $mg\,L^{-1}$, pH 7, and 40% CD removed 99.6% Chl-a and reduced 94.1% turbidity. The optimal removal efficiencies of Chl-a by PADB1, CPAM, PFS, and PAC were 99.1, 97.3, 84.6, and 65.3% at dosages of 3.5, 3, 35, and 30 $mg\,L^{-1}$, respectively. Turbidity and Chl-a removal by PADB resulted in charge neutralization and adsorption–bridging.

(2) The results of the fractal dimension analysis showed that the floc structures had strongly similar fractal characteristics. Fractal dimension of the formed flocs increased with increasing pH. High dosage corresponded to large fractal dimensions.

(3) Comparison results showed that the flocculation efficiency of PADB was significantly better than those of commercially available flocculants (CPAM, PFS, and PAC). PADB effectively removed Chl-a after flocculation, indicating the potential of PADB in treating algae-containing raw water.

Acknowledgements

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