Conductive Polyaniline Enhanced Decolorization of Azo Dyes in Anaerobic Wastewater Treatment

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Abstract

The polyaniline (PANI) was synthesized by chemical oxidation and was conductive media to improve the anaerobic sludge to decolorize the azo dye-containing wastewater in this study. The effect of carbon sources, chemical oxygen demand (COD) concentration and PANI dosage on the decolorization and degradation performance of anaerobic sludge was investigated. The optimized carbon source for decolorization of azo dye by anaerobic sludge was the sucrose in our research. The color removal can be increased by ~20% from the anaerobic system adding with PANI material due to the accelerated degradation of the carbon source by the PANI. Based on the decolorization, the azo dye-containing wastewater treatment could achieve an optimal effect when PANI of 400 mg/L was added in the anaerobic sludge. Finally, the effect of the additional PANI on the microorganism communities in the anaerobic sludge was also investigated.

Keywords: Azo dye; Polyaniline; Biological decolorization.

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1. Introduction

Azo dyes are widely used in textile, printing and dyeing, pharmaceutical and food industries due to their easy synthesis, rich color, stable chemical structure and low cost.^[1-2] During the production and utilization processes, a large number of azo dye-containing wastewater was produced, and became as the main type of dye wastewater in the recent years.^[3] This wastewater is always highly colored, thus it will affect the light transmittance of natural water bodies to inhibit photosynthesis of aquatic plants, thereby leading to the decrease in the dissolved oxygen in the water body and the death of other organisms in the water body.^[4-5] Moreover, the azo dyes have a high toxicity on human and environment, and have the risk of teratogenicity.^[6-7] Therefore, effective treatment of azo dye-containing wastewater has been widely concerned.

Azo dye molecules usually contain one or more azo bonds, thus the chemical oxidation and biological methods are often used to break the azo bonds to achieve the decolorization and

degradation of azo wastewater.^[8-9] Compared with the chemical oxidation methods, the biological treatment methods have widely used for treating this wastewater due to its low cost and environmental friendliness,^[10-11] especially the anaerobic biological method. Under anaerobic condition, the organic substrates in the wastewater are often degraded by the bacteria through the hydrolysis and acidification processes, generating the volatile fatty acids and the reductive bio-electrons in the anaerobic system.^[12] These electrons are transferred diffusively to reduce the azo bonds in the dye molecules, achieving the decolorization and degradation of the azo dye.^[13] The colorless aniline intermediates produced after the decolorization can be further mineralized by the bacteria under the anaerobic or aerobic condition. However, the anaerobic wastewater treatment method often has a long hydraulic retention time. Therefore, improving the efficiency of the microorganisms on the decolorization and degradation of the azo dyes has been considered as one of the challenges for treating this wastewater using the anaerobic biological method. It was supposed that accelerating the electrons transfer in the anaerobic sludge seems to be a relative way to improve the reduction of the azo bonds and degradation of the toxic intermediates.

Nowadays, the direct interspecies electron transfer process has been demonstrated as a highly effective way of the electron transfer between the microorganisms. In order to

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improve the efficiency of interspecies electron transfer, various artificial materials have been proven as the conductive medium to add into the anaerobic systems, including the active carbon, iron-based materials and conductive polymers. In the direct interspecies electron transfer process, the microorganisms transfer the electrons from the relevant electroactive proteins on their cell membranes to the surface of the solid electron acceptor, which prevent electron transfer by the carriers such as hydrogen through diffusion.^[14-15] As was known, the efficiency of electron transfer through the conductive materials is higher than that by the diffusion transfer of hydrogen gas, as the hydrogen diffusion in the anaerobic sludge was derived by the concentration difference.^[16]

Among the various conductive materials, the polyaniline (PANI) is a kind of conductive polymer containing benzene structure unit and quinone structure unit.^[17-18] It has been widely used in various applications due to its good electrical conductivity, stability and environmental friendliness.^[19-21] The surface defect of the PANI facilitates the electron transfer at its interface. When the PANI get the electrons, the surface charge density of PANI will change accordingly. As the charge can be conjugated, the electrons can be transferred along the molecular chain of PANI to adjacent atoms.^[22] With the help of conjugated π electrons, the electrons can be moved on the entire molecular chain of PANI.^[23] In the previous work, the PANI has been used as the conductive mediates to accelerate the electron transfer in the anaerobic wastewater treatment, thus improving the methane production from organic matters. It was supposed that the transferred electrons can be used for the reduction of the azo bonds to decolorized the azo dyes-containing wastewater.

In this study, the PANI was synthesized and was used as the conductive media to enhance the anaerobic sludge to decolorize and degrade the azo dyes in the wastewater. The influence of factors such as the carbon sources, COD concentration, and the PANI dosage on the decolorization and degradation of the azo dyes-containing wastewater was investigated. Then the mechanisms involved in the improved decolorization of anaerobic sludge by the PANI was disclosed with the microorganisms communities.

2. Materials and methods

2.1 Materials

Cationic Red X-GRL (X-GRL), a typical azo dye used in this study, were obtained by Guangzhou Jiheng Pharmaceutical Technology Co., Ltd., China. The sucrose, sodium acetate, sodium propionate were purchased from Shanghai Sinopharm Co. China. Aniline (C_6H_7N , 99.5%) were obtained by Sinopharm chemical Reagent Co., Ltd. Hydrochloric acid (HCl, 37%) and ammonium persulfate (APS, $(NH_4)_2S_2O_8$, 98%) were purchased from Beijing chemical works, China. All chemicals were used directly without further treatment. The anaerobic activated sludge was gained from a laboratory-scaled upflow anaerobic sludge bed, which treated wastewater

containing X-GRL.

2.2 Synthesis of PANI

The conductive PANI was synthesized by a chemical oxidation polymerization method.^[24-25] Dissolving 350 mM aniline monomer in 50 mL HCl solution (54 mM). The solution was sonicated for 20 minutes, then it was put into the ice-bath to keep the temperature at 0 °C. The mixture solution was stirred for 1h at 600 rpm, then 50 mL APS solution with a concentration of 350 mM was added to the mixture solution in a dropwise way. The mixed solution was kept at at 0°C and was stirred for 4 hours until the color of the solution turned dark green. The precipitated PANI particles was obtained by filtrating, and it was further washed with acetone and distilled water until the filtrate was colorless. The obtained PANI samples were vacuum dried at 60 °C for 12 hours.

2.3 Treatment of X-GRL wastewater

The anaerobic sludge was centrifuged at 3000 rpm for 5 minutes to remove the supernatant and resuspended in phosphate buffer, then it was suspended in the oxygen-free water again. 200 mL anaerobic sludge and 300 mL wastewater solution with a X-GRL concentration of 400 mg/L were added in a serum bottle. Then desired amount of PANI was mixed into the anaerobic systems. The wastewater solution (per liter) mainly contains carbon sources (including sucrose, sodium acetate, sodium propionate), KH_2PO_4 (20 mg), NH_4Cl (200 mg), $MgCl_2 \cdot 6H_2O$ (100 mg) and 2 mL trace elements solution. To ensure an anaerobic environment, the water used in this experiment was aerated with N_2 for 30 minutes to remove O_2 . Then all the bottles with and without added PANI were placed in the water with keeping the incubation temperature at 35°C. The sucrose, sodium acetate, sodium propionate were added in the nutrient solution which investigated the influence of the carbon sources on the decolorization. To investigate the impact of COD concentration, the COD concentration in the nutrient solution was controlled at 1000, 2000, 4000 mg/L, respectively. Also, the conductive PANI dosage was set as 100, 200, 400, 800 mg/L. Three parallel groups were conducted for each experiment. The wastewater was sampled for a desired time interval. The concentration of X-GRL was detected by a UV-Vis with setting the wavelength at 530 nm. The COD concentration was detected by the standard method using Lianhua digestion apparatus (China, LH-CM3H). The anaerobic sludges amended with and without conductive PANI were sampled, and microorganisms communities were detected by 16S rRNA high-throughput sequencing method.

2.4 Characterizations

The morphology of synthesized PANI was observed by a JEOL scanning electron microscope (JSM-7001F). The active groups of the PANI was measured by a Bruker Fourier infrared spectroscopy (FTIR, Vertex 70) with the a resolution of 4 cm^{-1} . X-ray photoelectron spectroscopy (XPS, PerkinElmer, PHI-5300/ESCA) was performed to characterize

the valence states of the N element in the conductive PANI, using monochromatic Al K α radiation.

3. Results and Discussion

3.1 Characterizations of PANI

The SEM was used to analyze the morphology and detailed structure of the synthesized conductive PANI (Fig. 1). It can be observed that the prepared PANI exhibit a regular rod-like structure with an average diameter of about 200 nm and length of about 1 μ m. It was also observed that the PANI have a rough surface, which is expected to make the microorganisms could easily adhere to its surface. The nano-sized PANI rod also facilitate the direct contact of the PANI with the microorganisms. The tight contact would shorten the electrons transfer distance between the bacteria and the conductive PANI, thus increasing the efficiency of electrons transfer between microorganisms in the anaerobic system.

The FT-IR spectra characteristics of the synthesized PANI have been shown in Fig. 2(A). The characteristic bands of PANI are observed at 1568 cm^{-1} , 1485 cm^{-1} , 1297 cm^{-1} , 1034 cm^{-1} and 792 cm^{-1} , which were corresponding to the quinoid C=C stretching, benzenoid C=C stretching, aromatic amine C-N stretching, quinoid ring -NH $^+$ - benzenoid ring stretching and C-H bending, respectively.^[26-27] The obvious peaks at 1568 cm^{-1} and 1485 cm^{-1} represent the -N= and -NH- in the PANI, which further indicated that the synthesized PANI existed as the redox intermediate state with a good electrical conductivity. Fig. 2(B) shows the N 1s XPS spectra of the synthesized PANI. The N 1s XPS spectrum can be deconvoluted to three main peaks located at 399.3 eV, 399.9 eV and 400.7 eV, which corresponded to benzenoid amine (-N=), quinoid amine (-N-) and protonated quinoid imine (-NH $^+$ =), respectively.^[28] As shown in the N 1s XPS spectrum, the ratio of the peak areas of the two characteristic peaks -N= and -N- is close to 1, which further indicated that the PANI synthesized in this study is in the intermediate redox state. It was also found that the prepared PANI was doped with protonated acids. These makes the PANI a highly conductive, which is important for facilitating the electrons transfer in the anaerobic sludge.

3.2 Effect of PANI on dye-containing wastewater treatment

The type of carbon sources is a key factor for the decolorization of X-GRL by microorganisms in anaerobic sludge, which act as the electron donors to provide electrons for X-GRL decolorization.^[29] In this work, the sucrose, glucose, acetate, and propionate were used as the carbon sources to add into the nutrient solution of the anaerobic system. The effect of the carbon sources on color and COD removal were measured (Fig. 3). It was found that the color removal percentage by the anaerobic systems added with the conductive PANI was obvious faster than the control groups, which indicated the improvement of the decolorization by the PANI. 96% of color removal percentage was achieved for the anaerobic systems using the glucose and acetate as the carbon sources, and 94.7% and 92.2% color was removed by the anaerobic sludge using the sucrose and propionate as the carbon sources, respectively. It was also found that the COD removal percentage in the control groups was significantly different with the groups added with the conductive PANI. COD concentration after treatment in the anaerobic systems added with PANI was lower than the control groups, which revealed that the conductive PANI also enhanced the COD degradation. This was consistent with the results reported in the previous works.^[30] As shown in the Fig. S1, the COD removal from the systems using the glucose and sucrose as the carbon source was faster than these using the propionate and acetate as carbon sources. Compared with the other two carbon sources, the glucose and sucrose are easily degraded by the anaerobic bacteria and release the electrons from the anaerobic sludge. The fast generation of electrons would reduce the chromophore groups, thereby achieving rapid decolorization.^[31] As shown in Fig. 3, most of the color was removed in the initial 20 minutes of incubation due to the fast generation of electron from the enough carbon sources. Moreover, the color removal percentages was increased by more than 20% by the anaerobic sludge added the PANI within the initial 20 minutes, it was inferred that the improvement of COD degradation by the conductive PANI attributed to the fast decolorization. Based on the performance

Fig. 1 SEM images of PANI at different magnifications.

Fig. 2 (A) FT-IR spectra and (B) N1s XPS of the synthesized PANI.

of color and COD removal, the sucrose was suggested as the candidate carbon source for the azo dye decolorization by using the anaerobic sludge.^[32] Thus the sucrose was selected as the carbon source for the further investigation.

To further explore the impact of the sucrose concentration on the X-GRL degradation by anaerobic sludge, 1000, 1500, 2000 and 4000 mg/L of sucrose were added to the anaerobic system. The color and COD removal were measured and showed in Fig. 4 and Fig. S2. The COD concentration remained in the effluent increased with increasing the initial COD concentration, and about 500 mg/L of COD was still remained in the effluent when then initial COD was higher than 2000 mg/L. Most of color was removed from the anaerobic systems in the initial 20 minutes, and the color removal percentage from the systems added with PANI was

higher than the control groups without adding the conductive PANI. 99.6%, 95.6%, 94.7% and 87.6% of color removal percentage were achieved when the initial COD concentration was 1000, 1500, 2000 and 4000 mg/L, respectively. The decolorization efficiency by anaerobic sludge was slightly reduced along with the increase in the initial COD concentration. It was possibly because the excessive sucrose led to the competition between quinone-respiring bacteria and methanogens for carbon sources, weakening the reduction of azo bonds and decolorization efficiency.^[33] This indicated that the improvement of the color and COD removal by the additional conductive PANI was also achieved in the anaerobic sludge feeding with different concentration of carbon source.

Fig. 3 Effect of carbon sources ((A) sucrose (B) glucose (C) acetate and (D) propionate) on decolorization of dye-containing wastewater of anaerobic sludge added with conductive PANI.

Fig. 4 Effect of initial COD concentration ((A) 1000, (B) 1500, (C) 2000 and (D) 4000 mg/L) on the decolorization of dye-containing wastewater.

Fig. 5 Effect of PANI dosage on the decolorization of X-GRL.

The conductive material polyaniline can theoretically be used as an electronic conductive medium to strengthen the anaerobic treatment of azo dyes and improve the decolorization of dyes. The material dosage is an important factor affecting the electron transfer in the anaerobic system.^[34] Fig. 5 showed the effect of PANI dosage on the decolorization of the X-GRL dye. It was also found that the PANI dosage obviously affect the reduction of azo bonds. As shown in Fig. 5, the X-GRL had been rapidly decolorized in the initial 20 minutes, and more than 80% of color was removed from the anaerobic systems added with 100, 200, 400 and 800 mg/L of

PANI. Moreover, the color removal percentage achieved by the anaerobic systems added with 400 mg/L of PANI, which was higher than the other systems. Excessive conductive materials may form aggregates with the extracellular polymeric substances secreted by microorganisms in the anaerobic sludge,^[35] which decreased the direct contact of the PANI materials with microorganism cells. On the other hand, too much nanomaterials will also has the toxic effect on the activity of the microorganism, decreasing the decolorization of the azo dyes in the wastewater. Thus, the optimal PANI dosage for the anaerobic sludge treating the azo dye-containing wastewater was 400 mg/L.

The microorganisms communities in the anaerobic system whether to add the conductive PANI were conducted. The addition of PANI significantly affected the bacterial community, which was shown in Fig. 6A & B. At phylum level, *Firmicutes*, *Chloroflexi*, *Bacteroidetes* and *Proteobacteria* were detected as the dominant bacteria for the anaerobic system whether to add the PANI. As documented, *Firmicutes* bacteria plays a important role in the anaerobic sludge community due to the ability of hydrolysis and hydrogenogenic acidogenesis,^[36] and it was also reported that *Firmicutes* bacteria could degradate the aromatic compounds.^[37] *Chloroflexi* and *Proteobacteria* also have the ability to degrade organic matters. *Chloroflexi* is a typical filamentous bacterium in sewage treatment process, which degrades polysaccharides and proteins.^[38] *Proteobacteria* has the ability to degrade complex carbon sources, and most of

Fig. 6 (A) Proportion of domain bacterial and (B) bacterial community composition, (C) Proportion of domain archaea and (D) archaeal community composition in anaerobic sludge before and after adding with PANI.

them are obligate and facultative anaerobes, which are widely found in anaerobic reactors.^[39] *Bacteroidetes* bacteria have the ability of extracellular electron transfer,^[40] which may be related to the methanogenesis process of in the anaerobic systems. These bacteria in the anaerobic sludge might take part in the direct interspecies electron transfer process in azo dye reduction. As shown in genus level, the dominant genera were *Clostridium* and *Longilinea*. *Clostridium* is a major hydrogenogens (Fig. S3), which usually involves in methanogenesis process.^[41] *Longilinea* is a group of bacteria that ferment polysaccharides and proteins under strict anaerobic conditions.^[42] The two genera were related to the hydrolysis and fermentation of methanogenesis, which indicated that PANI had no significant effect on the hydrolysis and fermentation stage of methanogenesis. In addition, *Pseudomonas* and *Desulfovibrio* had been enriched to a certain extent after adding PANI. *Pseudomonas* and *Desulfovibrio* was reported to have the ability on degradation of azo dyes,^[43,44] which may be related to increased anaerobic degradation of azo dyes with adding the conductive PANI.

Fig. 6C & D showed the archaeal community of anaerobic sludge with and without adding PANI. The two genera, *Methanosaeta* and *Methanobacterium*, were detected as the dominant archaea in the both anaerobic sludge. *Methanosaeta* is acetoclastic methanogens that produces CH_4 using acetate as substrate.^[45] *Methanobacterium* is hydrotrophic methanogens that converts CO_2 and H_2 into CH_4 .^[46] This indicated that the acetoclastic methanogenesis was main pathways for methane production in anaerobic sludge. The addition of PANI changed the abundance of various genera of methanogens. The abundance of *Methanosaeta* and *Methanobacterium* declined, while the abundance of *Methanolinea* and *Methanomassiliicoccus* increased when the

conductive PANI added into the anaerobic system. *Methanolinea* is also hydrotrophic methanogens.^[46] *Methanomassiliicoccus* is hydrotrophic and methylotrophic methanogens, which reduces methyl to produce methane through restricted other methanogenic pathway.^[47] Beside the acetoclastic methanogenesis, *Methanosaeta* archaea were reported to be capable for direct interspecies electron transfer process in the anaerobic sludge.^[48] As shown in Fig. 6C & D, the abundance of *Methanosaeta* archaea slightly decreased in the anaerobic system when the PANI was added, but its proportion still accounted for 74%. It was inferred that the direct interspecies electron transfer process might also existed in the anaerobic sludge for reduction of azo dyes. The aromatics and azo dyes had toxic effect, which might highly inhibit on the activity of acetoclastic methanogens.^[49] This might lead to the slightly increase in the proportion of the hydrotrophic methanogens in the PANI amended sludge.

4. Conclusion

The PANI synthesized by chemical oxidation has a nano-rod shape and exists as a conductive state. And the PANI was demonstrated to have enhancement for decolorization and degradation of the azo dye-containing wastewater by anaerobic treatment. When the sucrose was used as the carbon source, the color removal efficiency of the anaerobic sludge can be increased by ~20% with adding 400 mg/L PANI. The increased degradation efficiency of the carbon source was the main contribution for the improved decolorization of the sludge adding with conductive PANI. Meanwhile, the addition of PANI can obviously affect the microbes communities in the anaerobic systems, which was also the main mechanism involved in the enhanced performance of anaerobic sludge on the decolorization and degradation by the conductive PANI.

Conflict of interest

There are no conflicts to declare.

Supporting information

Applicable.

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