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Bio-Fenton-Assisted Biological Process for Efficient Mineralization of Polycyclic Aromatic Hydrocarbons from the Environment

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Abstract: The intensive production of fossil fuels has led to serious polycyclic aromatic hydrocarbon (PAH) contamination in water and soil environments (as PAHs are typical types of emerging contaminants). Bio-Fenton, an alternative to Fenton oxidation, which generates hydrogen peroxide at a nearly neutral pH condition, could ideally work as a pretreatment to recalcitrant organics, which could be combined with the subsequent biological treatment without any need for pH adjustment. The present study investigated the performance of a Bio-Fenton-assisted biological process for mineralization of three typical types of PAHs. The hydrogen peroxide production, PAH removal, overall organic mineralization, and microbial community structure were comprehensively studied. The results showed that the combined process could achieve efficient chemical oxygen demand (COD) removal (88.1%) of mixed PAHs as compared to activated sludge (33.1%), where individual PAH removal efficiencies of 99.6%, 83.8%, and 91.3% were observed for naphthalene (NAP), anthracene (ANT), and pyrene (PYR), respectively, with the combined process.

Keywords: Bio-Fenton; bio-remediation; PAHs; microbial population

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1. Introduction

The development of modern industrialization has led to the accumulation of several types of persistent organic pollutants (POPs). Among them, polycyclic aromatic hydrocarbons (PAHs) are receiving more attention due to their carcinogenicity, mutagenicity, and teratogenicity [1–5]. Due to the high chemical stability of the benzene ring structure, nature degradation (mainly driven by environmental microorganisms) occurs at a very slow rate, resulting in its long persistence in the open environment [6–9]. To effectively remove PAHs from the environment, a variety of remediation technologies have been applied, such as biodegradation, thermal remediation, chemical oxidation, etc. Among them, biodegradation is the most economical option; it requires a long remediation period, which limits its application in the contaminated site remediation [10–12].

Advanced oxidation processes (AOPs) have been widely applied as tertiary treatments in wastewater engineering owing to their capability in degrading recalcitrant organic pollutants (including antibiotics) from wastewater with the help of reactive oxygen species (ROS) [13–16]. Among the AOPs, the Fenton reaction has been a popular option for decades due to its ease of operation and high redox potential. However, there are a few drawbacks to the Fenton reaction, such as acidic reaction pH and relatively high operation costs (similar to other AOPs), which hinder its application in some specific wastewater treatment cases. Bio-Fenton, an alternative to the Fenton reaction, which generates hydrogen peroxide at a nearly neutral pH condition, has overcome the limitation of the Fenton reaction [17,18]. With help of glucose oxidase (GOD), glucose could be oxidized to gluconic acid and hydrogen peroxide, where the chelating effect of gluconic acid facilitates the Bio-Fenton reaction to be performed under a neutral pH level, and the generated hydrogen peroxide could effectively oxidize the organic pollutants [19–21]. According to the existing literature, the Bio-Fenton reaction has proven its efficacy in degrading several types of recalcitrant organics, such as trichloroethylene (TCE), dyes, PAHs, etc. [22–25].

From an engineering point of view, cost-effectiveness is always of great concern for the application potential of any wastewater treatment process [26,27]. Regarding organic reduction, it is known that biological wastewater treatment is one of the most economical options in wastewater treatment. Owing to the neutral pH condition, Bio-Fenton could ideally work as a pretreatment of recalcitrant organics, combined with the subsequent biological treatment, without any need for pH adjustment. To assess the potential of this combined process, this study investigated the performance of a combined Bio-Fenton with an activated sludge process in treating a PAH mixture solution. The hydrogen peroxide production, PAH removal, overall organic mineralization, and microbial community structure were comprehensively studied. The results of this work may offer new insight into a cost-effective hybrid process for the efficient treatment of recalcitrant organics.

2. Materials and Methods

2.1. PAH Preparation

In this study, three typical PAHs, namely naphthalene (NAP), anthracene (ANT), and pyrene (PYR) were purchased from Sigma-Aldrich under an HPLC grade (>95%). Regarding the Bio-Fenton oxidation experiment, individual PAHs with concentrations of 50 mg/L each were prepared. For a batch experiment involving mixed PAH removal, all three kinds of PAHs with concentrations of 50 mg/L each were prepared, and a trace element solution was added to ensure the proper functioning of activated sludge. The PAH concentrations were measured using HPLC-MS (Shimadzu, Japan), according to a previous study [8].

2.2. Bio-Fenton Reaction

Bio-Fenton oxidation was performed in 500 mL glass beakers with magnetic stirring of 150 RPM to ensure efficient mixing. Glucose oxidase (10 U), iron (III) citrate (0.1 mM), and glucose (2 mM) were purchased from Sigma-Aldrich and prepared for Bio-Fenton. To understand the effect of pH on ROS generation, three initial pH conditions of 5.0, 6.0, and 7.0 were investigated for process optimization. In addition, an individual PAH degradation experiment was performed by adding 50 mg/L of antibiotics into a Bio-Fenton mixture solution under optimized pH. During the above 48 h of experiments, samples were taken for analyzing pH, H_2O_2, COD, and antibiotic concentrations. Specifically, an H_2O_2 concentration was determined using an Amplex Red hydrogen peroxide/peroxidase assay kit according to the manufacturer's instruction, while oxidation-reduction potential (ORP) was recorded via a portable multimeter (Hach, USA).

2.3. Batch Experiment for Antibiotic Removal

A PAH mixture solution with the addition of trace elements was prepared for a batch experiment involving the removal of antibiotics. Bio-Fenton was performed with the same experimental setup as described in Section 2.2. The biological treatment experiment was performed using a 500 mL glass beaker with micropore aeration to maintain a dissolved oxygen level between 2.0 and 3.0 mg/L. Activated sludge was collected from an aeration tank of a local municipal treatment plant, and mixed liquor suspended solids (MLSS) was adjusted to approximately 3000 mg/L for the biological PAH removal experiment. For the batch experiments, three types of PAHs were treated individually with initial concentrations of 50 mg/L by activated sludge. Subsequently, two parallel tests were conducted treating the PAH mixture: one was performed using only activated sludge for 120 h, while the other was performed using Bio-Fenton pretreatment for 24 h, which was followed by activated sludge treatment for another 120 h.

2.4. Microbial Community Analysis

After 120 h of experimenting, the activated sludge sample from each test was taken, and microbial population distributions among the different samples were compared. The DNA was extracted with an UltraClean DNA extraction kit (Mobio Laboratories, Carlsbad, CA, USA) according to the manufacturer's instructions. The 16S rRNA gene fragments from the total DNA of the sample sets were amplified using DreamTaq Green PCR Master Mix (Thermo Scientific, USA), with a primer set: 343F (5′-TACGGRAGGCAGCAG-3′)/926R (5′-CCGTCAATTYYTTTRAGTTT-3′). Raw sequences from pyrosequencing were screened; sequences less than 200 bp or containing ambiguous bases were excluded. The taxonomic identities of the sequences were then assigned using the Classifier program of the RDP-II, with a minimum confidence level of 80% [4].

3. Results and Discussion

3.1. Effect of pH on H_2O_2 Generation

The generation of H_2O_2 is the key parameter in the evaluation of oxidation potential during the Bio-Fenton reaction, while the reaction pH may determine its compatibility with a biological treatment, which could greatly reduce the overall treatment cost. As shown in Figure 1a, for all three pH conditions, H_2O_2 generation almost stabilized from 12 h, with concentrations of 203, 225, and 215 mg/L under initial pHs of 5.0, 6.0, and 7.0, respectively. The H_2O_2 concentrations tended to decrease slightly with an increase in the reaction time, which could have resulted from the instability of H_2O_2. The H_2O_2 concentration generated in the present study was consistent with the literature, from which, with a number of recalcitrant organics, including TCE, PAHs could be effectively degraded [13,19]. In addition, as shown in Figure 1b, during the Bio-Fenton reaction, the pH of the mixture solution dropped slightly, which might have been caused by the transformation of gluconic acid from glucose. Considering the observation that pH did not show a significant impact on H_2O_2 generation, and neutral pH is known to be more favorable for biological treatment as well as more friendly to the receiving environment, the initial pH of 7.0 was determined as an optimal pH for the subsequent experiments.

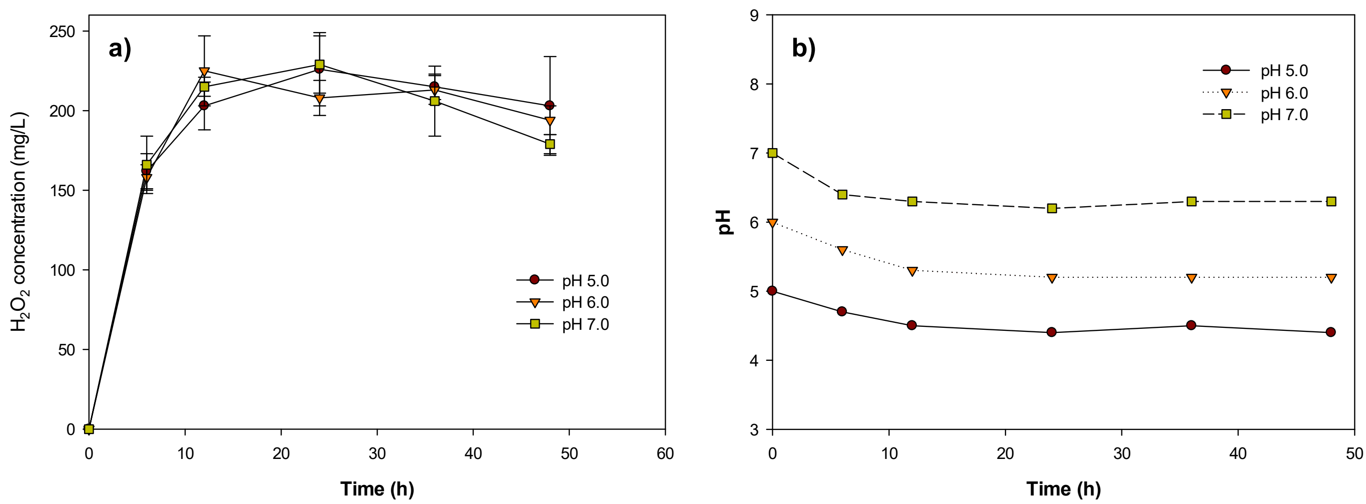

Figure 1. Effect of pH on H_2O_2 generation: (**a**) H_2O_2 generation with different initial pHs; (**b**) pH change with reaction time.

3.2. Bio-Fenton Pretreatment for PAH Removal

The Bio-Fenton oxidations on three individual PAHs were assessed, based not only on PAH removal but also on the overall organic mineralization (COD removal). As illustrated in Figure 2a, within 48 h, varying overall COD removal efficiencies were observed from Bio-Fenton oxidation, with values of 95.1%, 75.4%, and 85.2% for NAP, ANT, and PYR, respectively. Similar findings have reported that efficient PAH removal could be observed under various AOP treatments [1]. However, as shown in Figure 2b, much lower COD removal performances were observed through AOP oxidation, with removal rates of 28.6%, 13.8%, and 30.8% for NAP, ANT, and PYR, respectively, suggesting that, instead of complete mineralization, Bio-Fenton broke down the PAHs into smaller organic fractions, which had to be further treated to meet a satisfied COD removal or COD discharge limit. Moreover, to investigate the feasibility of a subsequent biological treatment, the H_2O_2 residue concentration, as well as ORP during Bio-Fenton oxidation, were measured and illustrated (see Figure 2c). As can be seen, for all three PAHs, the H_2O_2 residue in the mixed liquor followed an initial increase with a gradually decreasing trend, which could be explained by the balance of H_2O_2 generation during Bio-Fenton and H_2O_2 consumption during PAH removal. At the end of the experiment (48 h), the H_2O_2 residue concentrations ranged at relatively low levels between 1.0 and 23.0 mg/L. The ORP values, in particular, ranged between 231 and 305 mV, which were considered typical ORP values for an aerobic process [5], suggesting a relatively low or non-inhibitory mixed liquor environment towards a subsequent activated sludge process.

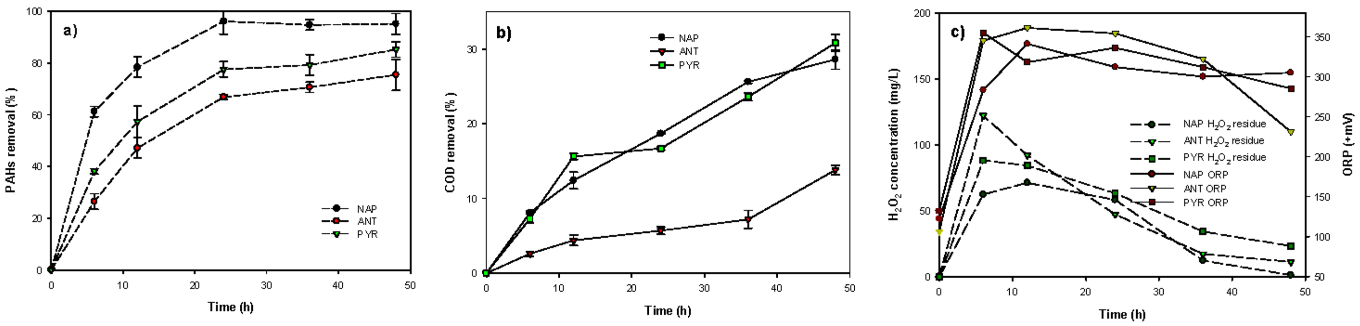

Figure 2. Bio-Fenton reaction for the three antibiotic degradations: (**a**) PAH removal; (**b**) COD removal; (**c**) H_2O_2 residue and ORP conditions during Bio-Fenton.

3.3. Combined Bio-Fenton with Activated Sludge for PAH Removal

Each PAH investigated in the present study is known for its inhibitory effects on microbial activity. Figure 3a shows the removal efficiencies of individual PAHs under activated sludge treatment. As can be seen, with 120 h of treatment, NAP could most effectively be removed (82.0%), followed by PYR (55.0%) and ANT (41.0%). These values indicate that among the three PAHs, two of them cannot be effectively treated (with removal rates $< 60\%$) through an activated sludge process, which could be explained by the relatively higher biotoxicity [3]. In addition, the COD removal performance comparison between only activated sludge and combined Bio-Fenton with activated sludge treating mixture PAHs was conducted (shown in Figure 2b). Only 33.1% of COD removal could be achieved using activated sludge, which suggests that the mixture of three types of PAHs significantly increases the bio-toxicity effect as compared with individual PAHs. Meanwhile, a combined Bio-Fenton with activated sludge could achieve remarkable COD removal of 88.1%, where individual PAH removal efficiencies of 99.6%, 83.8%, and 91.3% were observed for NAP, ANT, and PYR, respectively. To further understand the COD removal contribution (Figure 3c) for the combined process, although Bio-Fenton pretreatment only contributed 15.9% of COD removal, with its role in reducing the bio-toxicity, the performance of activated sludge could be improved by 118%, which could achieve 33.1% of COD removal without Bio-Fenton, and 72.2% of COD removal with Bio-Fenton. This finding strongly supports the assumption that Bio-Fenton could be a promising pretreatment technology to be coupled with a conventional biological treatment when handling recalcitrant organic wastewater [13].

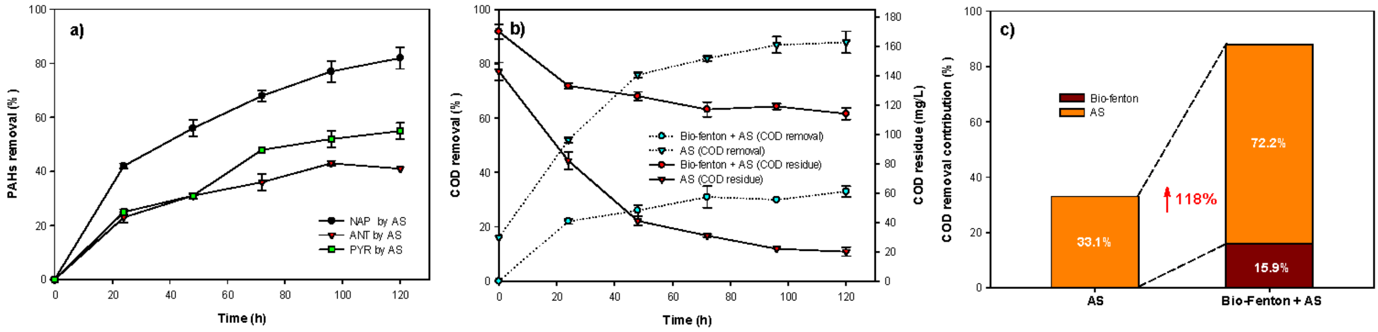

Figure 3. Combined Bio-Fenton with activated sludge for PAH removal: (**a**) PHA removal by activated sludge (AS); (**b**) COD removal by AS with/without Bio-Fenton; (**c**) COD removal contribution.

3.4. Microbial Community

The microbial population and community structure could 'reflex' the key functional microbial groups that are responsible for organic biodegradation. The top ten abundant microbial populations at the phylum level are summarized in Figure 4. In general, *Proteobacteria*, *Bacteroidetes*, *Acidobacteria*, *Chloroflexi*, and *Planctomycetes* were found to be the most abundant groups, accounting for 62.7–81.6% of the total species among the three sludge samples. Interestingly, without Bio-Fenton pretreatment, a more significant swift of the microbial community structure was found as compared to the one with Bio-Fenton, which indicated that the potential toxicity of PAHs was a great driving force for microbial community evolution by inhibiting the lower toxic-resistant species. In particular, among these ten phyla, *Proteobacteria*, *Acidobacteria*, and *Spirochaetes* were found to be remarkably decreased in their abundance with 120 h of exposure to PAH mixtures, while for *Bacteroidetes*, *Chloroflexi*, *Planctomycetes*, and *Firmicutes*, an opposite trend of an increase in abundance was observed [7]. This phenomenon suggested that with biotoxicity as a specific environmental stress, microbial acclimation might take place with microbial population evolution to achieve a new balance between higher toxic-resistant microbial consortia and the unfavorable environment. Moreover, the microbial population results also indicated that with the help of Bio-Fenton to reduce bio-toxicity through oxidation, it could be effective at improving the removal efficiency of the biological process when handling recalcitrant or toxic organic wastewater.

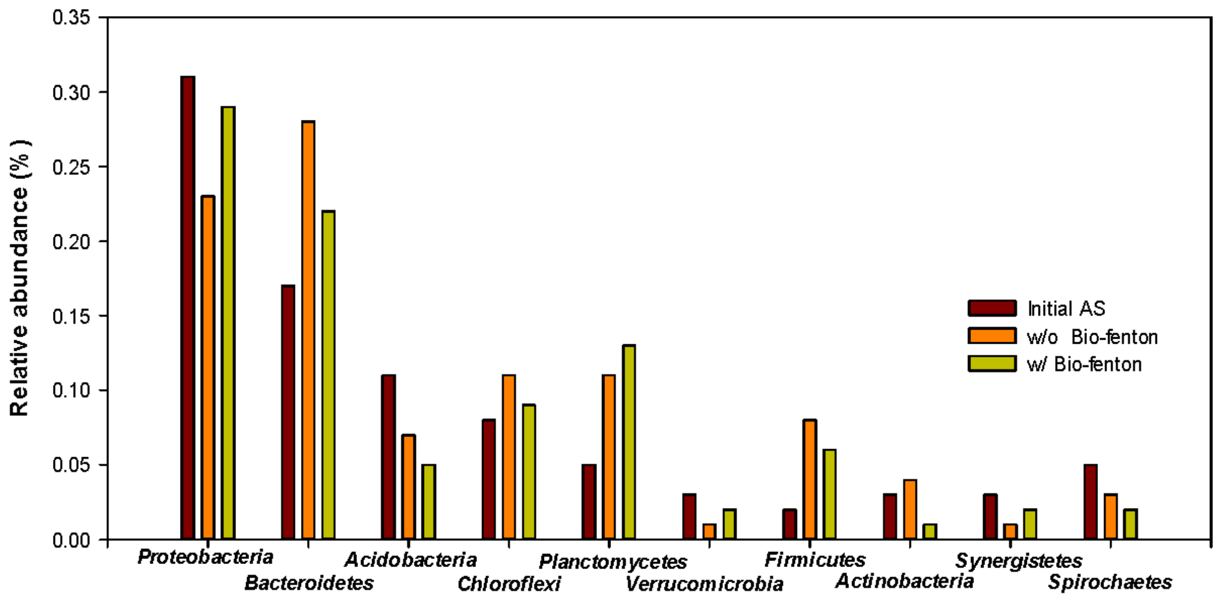

Figure 4. Microbial community structure in sludge samples.

4. Conclusions

In the present study, Bio-Fenton showed its potential as a pretreatment to be coupled with conventional activated sludge for treating PAH mixtures. Bio-Fenton could effectively break down individual PAHs ranging from 85.2 to 95.1%; with the help of Bio-Fenton, the coupled activated sludge could achieve COD removal of 88.1%, compared with only 33.1% for activated sludge alone. Regarding the microbial population, *Proteobacteria*, *Bacteroidetes*, and *Acidobacteria* were the predominant genera in the sludge samples.

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