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Correspondence analysis of bio-refractory compounds degradation and microbiological community distribution in anaerobic filter for coking wastewater treatment

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HIGHLIGHTS

- Coking wastewater was efficiently treated by I-AF and I-BAF.
- Correlation between bio-refractory compounds degradation and microbial community was explored.
- *Enterobacter*, *Pseudomonas* and *Sedimentibacter* related to phenol degradation.
- *Enterobacter and Sedimentibacter* related to methylphenol degradation.
- *Sedimentibacter* was firstly found related to many compounds, e.g. naphthol etc. degradation.

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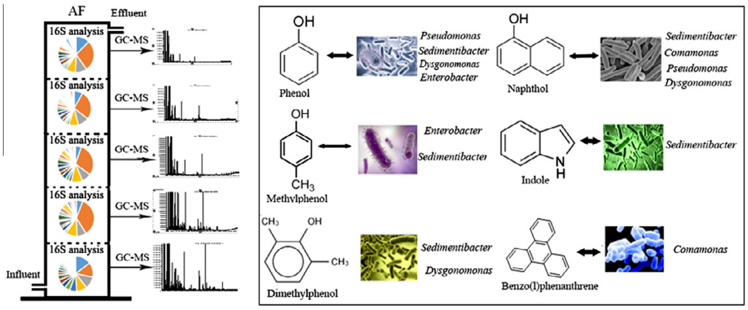
GRAPHICAL ABSTRACT



ABSTRACT

Coking wastewater usually contains lots of mutative bio-refractory compounds, which causes the difficulties for the biological coking wastewater treatment. In this work, we employed Gas Chromatography–Mass Spectrometer to determine organic compounds concentration along different heights of anaerobic filter and 16SrRNA high-throughput gene sequencing analysis to reveal microbial community distribution. As results, 360 genera were detected in the reactor, where *Acrpbacter*, *Proteiniphilum*, *Pseudomonas*, *Comamonas*, *Sedimentibacter*, *Enterobacter* and *Dysgonomonas* dominated the microbiological community. They were found to have obvious correlations with the degradation of some bio-refractory compounds including phenol, methylphenol, dimethylphenol, naphthol, triphenylene and indole in coking wastewater. Specially, phenol degradation has a high coefficient with *Enterobacter*, *Pseudomonas* and *Sedimentibacter*. Methylphenol degradation exhibits the good relativity with *Enterobacter and Sedimentibacter*. Besides the well-known *Enterobacter*, *Dysgonomonas*, *Pseudomonas* and *Comamonas* for phenol and methylphenol degradation, most importantly, we found the high correlation coefficients of *Sedimentibacter* to phenol, methylphenol, dimethylphenol and naphthol degradation. The new finding would be benefit for the identification of *Sedimentibacter* for phenol, methylphenol, dimethylphenol and naphthol degradation, and the isolation of related bacteria for bio-augmentation. This study is useful to reveal "black box" model of coking wastewater biological treatment and provides another way to investigate functional bacteria to degrade bio-refractory compounds.

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1. Introduction

As one of the most general fuel and chemical feedstock in China, coal is widely used in various factories and industries [1,2]. Coking wastewater could be produced during the industrial processes of coal coking, coal gas purification, liquefaction and refining [3]. Some contaminants such as thiocyanate, cyanide, polynuclear aromatic hydrocarbons (PAHs) and nitrogenous heterocyclic compounds are bio-refractory organics, which are widely considered to be carcinogenic and mutative. They have been reported as main contaminants in coking wastewater [1,3].

Among all treatment methods for degrading toxic organics in coking wastewater, biological process is deemed to be efficient, affordable and has the least environmental and health risk [2]. Biological treatment processes such as anaerobic–anoxic–oxic system and anoxic–oxic system can achieve high removal efficiency of chemical oxygen demand (COD), nitrogen, cyanide, thiocyanate, phenols, polycyclic and heterocyclic compounds [4–7]. Meanwhile, the stability of biological treatment stages is adversely affected by the high load of toxicity of the compounds [4]. Therefore, it is agreed that bacteria strains with high activity and robust physiology are critical to the stability of biological treatment process because they can efficiently facilitate the removal of refractory organics, mitigate wastewater toxicity and thus improve wastewater biodegradability [4,7–9].

Many studies have focused on the treatment of coking wastewater containing various bio-refractory organics by high-efficiency bacteria strains. For instance, researchers reported that bacteria strains *Burkholderia pickettii* could enhance the removal of quinoline from coking wastewater in anaerobic–anoxic–oxic system [1]; *Comamonas* was discovered to facilitate the resistance of activated sludge reactor to 3-chloroaniline shock load [10]. However, the previous studies mostly focused on the removal of certain organics such as phenolic [6,11], cyanide, thiocyanide [12], PAHs [7] and N-heterocyclic compounds [13] by some high-efficient bacteria strains. Few studies investigated the further correlation between organics degradation and microbial community, which is always regarded as a “black box” model.

In this study, one Anaerobic Filter-Biological Aerated Filter (AF-BAF) system was set up. Reactor parameters, such as Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD_5), NH_4^+-N and pH, were measured during different treatment phases. Moreover, we got the water and sludge samples from the different heights of the AF reactor, and applied Gas Chromatography–Mass Spectrometer (GC–MS) to detect the compounds concentration, high-throughput 16SrRNA gene sequencing analysis to analyze the microbial community at different heights of the reactor. Subsequently, we set up the relationship between the degradation of organics and the distribution of microbial community, and then pointed out the kinds of bacteria which are largely responsible for the degradation of a certain contaminant.

2. Materials and methods

2.1. Raw wastewater

Raw wastewater was collected from a coking factory located in Shanxi Province, the most famous coal producing area of China. The wastewater with deep brown color and obnoxious odor was collected without any pretreatment. Ammonium, phenols and oil content were all slightly higher compared to other reported studies [3,6]. The concentrations of main parameters in influent are listed in Table 1. A dosage of 0.05 g KH_2PO_4/L was added as phosphorous resource for microorganism growth. Concentrated sulfuric acid was added into the influent to adjust pH to 7.5–8.5.

Table 1
Characteristic of influent of AF-BAF system.

Parameter	Concentrations (mg/L)
COD	1400–2300
BOD	740–800
V-phenols	380–600
NH_4^+-N	260–400
NO_2^--N	ND
NO_3^--N	2–10
SS	120–350

2.2. Filter reactors

The bench-scale AF-BAF system (Fig. 1) consisted of an anaerobic reactor (AF, working volume of 4.5 L) and an aerobic reactor (BAF, working volume of 4.5 L). Two reactors were all made of polymethyl methacrylate. Six ports were installed at heights of 0 cm (at bottom of the reactor, near the influent inlet), 10 cm, 20 cm, 30 cm, 40 cm and 50 cm respectively for collecting water samples at day 250 in AF reactor. Five sludge samples were harvested at locations of 0–10 cm, 10–20 cm, 20–30 cm, 30–40 cm and 40–50 cm at the same day (day 250). Carriers that used in reactors were self-made patented Functional PolyUrethane FoamS (FPUFS). They have been successfully applied in other toxic and refractory wastewater treatment system [15,16]. The carriers with size of 50 mm × 50 mm × 50 mm, density of 1.0 g/cm^3 and specific surface area of 35,000 m^2/m^3 were carefully placed without compression into the reactor under dry condition.

The temperature of the reactors was kept at 35 ± 2 °C with a water-bath, which is close to the temperature range in real coking wastewater treatment facilities [2]. Dissolved Oxygen (DO) of the AF reactor was kept below 0.1 mg/L and air diffusers were used to maintain DO around 4–6 mg/L in BAF reactor. HRT was kept at 20 h during the phase I, and 32 h during the phases II, III and IV of the reactor operation. Correspondingly, the volumetric flow of the reactor was 0.23 L/h during the phase I, and 0.14 L/h during the phases II, III and IV. The whole reactor operation lasted for 250 days.

2.3. Inoculated microorganism

One group of microorganisms B350, purchased from BIO-SYSTEMS Co. (USA), was added in both reactors. B350 has been proved to have high efficiency in the treatment of oil field wastewater [15]. No sludge discharging was performed during the reactor operation.

2.4. Chemical analytical methods

2.4.1. Wastewater parameters detection

COD, NH_4^+-N, Total Nitrogen (TN), NO_3, and V-phenols were measured using Standard Methods. BOD_5 was monitored using respirometric method by OxiTop bottles (WTW Co., Germany). A pH meter (Mettler Co., Switzerland) was used to identify pH in each reactor and DO was detected by a DO meter (Mettler Co., Switzerland).

The reactor loading was calculated according to the following formula:
Reactor loading rate = Con.inf/HRT
The reactor removal rate was calculated according to the following formula:
Reactor removal rate = (Con.inf − Con.eff)/HRT
The reactor removal efficiency was calculated according to the following formula: