



Removal and fate of polycyclic aromatic hydrocarbons in a hybrid anaerobic–anoxic–oxic process for highly toxic coke wastewater treatment

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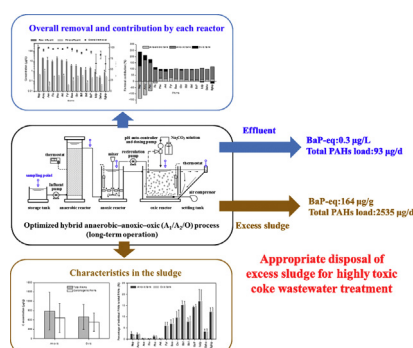
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HIGHLIGHTS

- High concentrations of PAHs were observed in the raw coke wastewater.
- PAHs were significantly removed (99%) by the optimized hybrid $A_1/A_2/O$ process.
- Activated sludge treatment was crucial for PAHs elimination through adsorption.
- The daily load and BaP-eq of PAHs were extremely high in the excess sludge.
- Appropriate disposal of excess sludge for coke wastewater was highlighted.

GRAPHICAL ABSTRACT



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ABSTRACT

Elimination of polycyclic aromatic hydrocarbons (PAHs) from coke wastewater is crucial to minimize the PAHs contamination levels to the environment. Knowledge about the characteristics of PAHs removal in biological treatment processes, especially hybrid systems, for real coke wastewater treatment has been very scarce. In this study, a lab-scale hybrid anaerobic–anoxic–oxic ($A_1/A_2/O$) process was used to treat highly toxic coke wastewater and operated more than 600 d at total hydraulic retention time (HRT) of 50 h, 40 h, 30 h, 20 h and internal mixed liquor recirculation ratio (R) of 3, 6, 9. Removal performance and behaviors of priority PAHs in the hybrid $A_1/A_2/O$ system were investigated. The results showed that the appropriate total HRT and R from oxic reactor to anoxic reactor for organics and nitrogen removal was 40 h and 3, respectively. The concentrations of total PAHs were very high (254–488 $\mu\text{g/L}$) in the raw coke wastewater, and effectively reduced to 4.1–4.5 $\mu\text{g/L}$ in the final effluent by the present system under the optimized operational conditions. Among the three treatment units, anoxic reactor made the largest contribution to the total PAHs removal. Large amounts of PAHs (415–1310 $\mu\text{g/g}$) were adsorbed to the activated sludge in the anoxic and oxic reactor, leading to a much higher load of PAHs (2535 $\mu\text{g/d}$) in the excess sludge than that in the treated coke wastewater (93 $\mu\text{g/d}$) at SRT 60 d. Therefore, the excess sludge was identified as the major emission source of PAHs in coke wastewater during the hybrid $A_1/A_2/O$ process, and might pose an environmental risk if the excess sludge was not properly treated and disposed.

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1. Introduction

Coke wastewater, generated from coal carbonization and fuel classification processes in the iron and steel industry, contains high concentration of ammonia, cyanide, thiocyanate and phenols, as well as certain quantities of polycyclic aromatic hydrocarbons (PAHs) and was regarded as one of the most toxic industrial effluents (Zhao et al., 2009a; Zhang et al., 2012). This heavily loaded and hazardous wastewater can induce severely adverse effects to the environment without properly treatment (Kim et al., 2008; Sharma and Philip, 2016; Huang et al., 2016; Joshi et al., 2017).

Biological treatments principally treat the coke wastewater. Among various proposed biological processes, the anaerobic-anoxic-oxic ($A_1/A_2/O$) system, offering the anaerobic acidification stage and the simultaneous denitrification stage, preferably remove the refractory polycyclic and heterocyclic compounds (Zhao et al., 2009b; Na et al., 2017; Shi et al., 2017). Hybrid process, involving both biofilm and suspended biomass, can enhance the biodiversity in the system and improve the biodegradation of hazardous organics, compared to the conventional $A_1/A_2/O$ system (Lai et al., 2008; Zhou et al., 2017).

PAHs have been demonstrated as one of the most hazardous constituents in the coke wastewater (Na et al., 2017). However, knowledge about the removal of PAHs by the biological treatment processes, especially hybrid biological treatment systems, is very limited. In several recent studies about the treatment performance of coke wastewater, PAHs were only qualitatively analyzed (Ma et al., 2016) or could not be quantified in coke wastewater due to the high detection limits (Sharma and Philip, 2016). The lack of regulation for PAHs in the coke wastewater might be one of the reasons for the limited investigation. PAHs were not included in the national emission standard for coke wastewater in China before 2012. However, since more and more evidences demonstrated that industrial wastewaters, especially those from steel companies, were the important source of PAHs in the environment (Ren et al., 2015), there have been increasing requirement for further enhancement of coke wastewater treatment considering PAHs removal. In 2012, the Ministry of Environment Protection of China published a new emission standard of pollutants for coke chemical industry, in which the concentration limit of total PAHs and benzo[a]pyrene in the treated wastewater were set as 50 $\mu\text{g/L}$ and 0.03 $\mu\text{g/L}$, respectively. Since then, researchers have started to characterize PAHs in real coke wastewater treatment plants (Zhang et al., 2015). However, since the sampling campaign was only conducted for a few days, some random uncertainty might be brought in due to the complicated conditions in the real coke wastewater treatment plants. By installing a laboratory fixed film reactor, Janosz-Rajczyk et al. (2014) discussed the possibility and efficiency of PAHs removal from coke wastewater under anaerobic conditions, and identified the role of sorption/desorption process in the removal of high-molecular-weight PAHs. Huang et al. (2016) operated a bench-scale anaerobic filter (AF) - biological aerated filter (BAF) system for coke wastewater treatment for 250 d, and investigated the degradation profile of typical PAHs along different heights of AF reactor. Their studies indicated the reliability of a more controllable set-up to better elucidate the behavior and fate of PAHs during the coke wastewater treatment process.

In this study, a lab-scale hybrid anaerobic-anoxic-oxic ($A_1/A_2/O$) process was installed to treat highly toxic coke wastewater and was operated for more than 600 d. The operational conditions of total hydraulic retention time (HRT) and internal mixed liquor recirculation ratio (R) from oxic reactor to anoxic reactor were optimized for organics and nitrogen removal. Under the optimum operational conditions, the priority PAHs in aqueous phase and activated sludge of coke wastewater were quantitatively determined. The contribution by each reactor to the removal of PAHs, occurrence of PAHs in the sludge and the major emission sources were extensively discussed. Furthermore, the benzo[a]pyrene equivalent concentration (BaP-eq) was employed to indicate the potential risk caused by PAHs released to the environment. The

results were expected to expand the understanding of the behavior of PAHs in coke wastewater during the optimized hybrid $A_1/A_2/O$ process, and help to reduce the release of PAHs to the environment from the source.

2. Materials and methods

2.1. Chemicals and materials

PAHs standard (EPA 610 PAHs mixture) including naphthalene (Nap), acenaphthylene (Acny), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo[a]anthracene (Baa), chrysene (Chr), benzo[b]fluoranthene (Bbf), benzo[k]fluoranthene (Bkf), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (Icdp), dibenzo[a,h]anthracene (Daha), benzo[g,h,i]perylene (Bghip), and deuterated internal standard containing acenaphthene-d10, phenanthrene-d10 and chrysene-d12 were purchased from Supelco (USA). The physicochemical properties of the 16 PAHs were listed in Table S1. HPLC-grade dichloromethane (DCM), acetone, hexane, ethyl acetate (EtAc) and methanol (MeOH) were provided by J.T. Baker (USA). Ultrapure water was produced by a Milli-Q water purification system (Millipore, France). Activated copper powder was prepared by washing with HCl, ultrapure water, MeOH, acetone, hexane successively, to remove surface oxide and impurity. Anhydrous sodium sulfate (Na_2SO_4) was dried at 400 °C for 4 h prior to use. Solid phase extraction cartridge ENVI-18 (6 mL, 1000 mg) was purchased from Supelco (USA). Neutral aluminum oxide (150 mesh) and silica gel (60–200 mesh) were obtained from Sigma-Aldrich (USA) and Acros Organics (USA), respectively. All the glassware was pre-washed and rinsed by MeOH and ultrapure water.

2.2. The reactors

The lab-scale hybrid $A_1/A_2/O$ system (Fig. 1) consisted of an upflow anaerobic reactor (working volume of 6 L, packed with a soft medium of hydrophilic acrylate fiber at 95% porosity), an anoxic reactor (working volume of 12 L, completely mixed) and an oxic reactor (working volume of 18 L, completely mixed).

A diaphragm pump (model X068-XB-AAAA365, Pulsafeeder, USA) fed the raw wastewater from a 100-L storage tank to the anaerobic reactor. The internal mixed liquor was recirculated from oxic reactor to anoxic reactor by a peristaltic pump (model NO.7520-57, Cole-Palmer, USA). The dissolved oxygen (DO) concentration of the oxic reactor was maintained >3 mg/L. The Na_2CO_3 solution was supplied to compensate alkalinity loss for nitrification and maintain pH in the oxic reactor at 7.0–7.2 by a pH controller (model Liquitron DP 5000, LMI MILTON ROY, USA) and electromagnetic dosing pump (model UL#A752-393SI, LMI MILTON ROY, USA). The average dosing flow rate of Na_2CO_3 was $<1.5\%$ of the influent feeding rate, hence minimizing its effects on the testing results. The three reactors were operated at 30–35 °C using thermostats (This temperature range was close to that in coke wastewater treatment facilities).

2.3. Wastewater and tests

Raw coke wastewater was collected bi-weekly from the Beijing Steel Company, China, and stored at 4 °C prior to tests. The wastewater was pretreated using ammonia stripping and gas flotation in the coke factory to decrease ammonia and oil concentrations. Phosphoric acid was added to adjust the pH to neutral and act as a phosphorus source for microorganisms. The characteristics of feeding coke wastewater were shown in Table S2.

The hybrid $A_1/A_2/O$ system was operated for >600 d. Table S3 lists the operation conditions. Briefly, the present system was successfully started up and subsequently operated stably for 108 d, and then different total HRT of 50 h, 40 h, 30 h, 20 h and R of 3, 6, 9 were tested to

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