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Simultaneous removal of aniline, nitrogen and phosphorus in aniline-containing wastewater treatment by using sequencing batch reactor

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HIGHLIGHTS

- A sequencing batch reactor was used to treat aniline-containing wastewater.
- Simultaneous removal of aniline, nitrogen and phosphorus was achieved.
- Good removal performance was maintained during variation of operating conditions.
- The accumulation of NH_4^+-N and TN during aniline biodegradation was alleviated.

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ABSTRACT

The high removal efficiencies of traditional biological aniline-degrading systems always lead to accumulation of ammonium. In this study, simultaneous removal of aniline, nitrogen and phosphorus in a single sequencing batch reactor was achieved by using anaerobic/aerobic/anoxic (A/O/A) operational process. The removal efficiencies of COD, NH_4^+-N, TN, TP were over 95.80%, 83.03%, 87.13%, 90.95%, respectively in most cases with 250 mg L^{-1} of initial aniline at 6 h cycle when DO was 5.5 ± 0.5 mg L^{-1}. Aniline was able to be completely degraded when initial concentrations were less than 750 mg L^{-1}. When DO increased, the removal rate of NH_4^+-N and TP slightly increased along with the moderate decrease of removal efficiencies of TN. The variation of HRT had obvious influence on removal performance of pollutants. The system showed high removal efficiencies of aniline, COD and nutrients during the variation of operating conditions, which might contribute to disposal of aniline-rich industrial wastewater.

1. Introduction

Aniline is hazardous to living beings and there has been more concern about its disposal along with extensive applications (Orge et al., 2015). Industrial wastewater, agricultural runoff and urban sewage often contain aniline and various other pollutants. Some of these compounds are toxic, carcinogenic, mutagenic and teratogenic. They are able to remain in water and soil for a long period of time, which has harmful impacts on environmental quality (Zhu et al., 2012). Therefore, it is important to treat wastewater containing these compounds before discharging into water bodies. China as well as USA has rated aniline as a persistent organic pollutant of which the release amount is strictly limited (Liu et al., 2015).

The dispose of aniline and most of its derivatives can be achieved by several physicochemical methods such as photodecomposition, electrolysis, ozone oxidation, resin adsorption and electro-Fenton, which have limits because of relatively high economic cost, energy consumption, and difficulty to remove pollutants completely (Qi et al., 2002; Anotai et al., 2006). Biological methods utilize microorganisms to thoroughly degrade aniline, which are suitable for large-scale wastewater treatment (Wang et al., 2007). Several biological technologies have been developed to remove aniline in recent years such as aniline-degrading bacteria, activated sludge reactors, moving bed biofilm reactors, which showed relatively high removal rates (Campos et al., 2002; Li et al., 2010; Dvořák et al., 2014). Most of these processes focus on realizing good removal performance of aniline and COD. However, the fact that accumulation of ammonia nitrogen (NH_4^+-N) and high concentration of total nitrogen (TN) coming from a large amount of nitrogen in aniline is always ignored in previous studies. Moreover, to our knowledge the phosphorus removal in these treatment systems has not meant mentioned.

Nitrogen and phosphorus are common pollutants in sewerage and industrial wastewater. Due to the fact that they are major

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causes of some environmental problems such as eutrophication and several diseases which affect human beings, their release has attached much attention. There have been increasing wastewater treatment plants over the world which are committed to removal of nitrogen and phosphorus (Li et al., 2014). Traditional nitrogen biodegradation consists of two main processes, namely nitrification and denitrification. Most of denitrifying bacteria are heterotrophic, which have a competitive relationship with phosphate-accumulating organisms (PAOs), leading to more difficulties in simultaneous removal of nitrogen and phosphorus (Meinhold et al., 1999). Unlike conventional phosphorus removal by release under anaerobic conditions and uptake in aerobic environments, denitrifying phosphate-accumulating organisms (DNPAOs) which belong to PAOs, have capability of uptaking phosphorus by nitrate and/or nitrite as electron acceptors in anoxic situation and utilizing O_2 under aerobic condition, resulting in denitrification and phosphorus removal at the same time (Zhang et al., 2011; Sun et al., 2015). Additionally, the process has less carbon source consumption as well as aeration costs and lower cell yield production, which makes DNPAOs have advantages over general biological methods to reduce nitrogen and phosphorus in wastewater (Wang et al., 2015a).

In the present study, DNPAOs was enriched in SBR reactor operated by anaerobic/aerobic/anoxic (A/O/A) mode and one of the two SBR reactors was fed with synthetic wastewater containing aniline. The effect of organic loading rate, aeration as well as HRT on aniline, COD and nutrients removal was evaluated and the variations of pollutants concentrations in two reactors within one cycle was investigated. To our knowledge, this is the first report of simultaneous removal of aniline, nitrogen and phosphorus in the single sequencing batch reactor. The work was conducted to remove both aniline and nitrogen coming from aniline biodegradation. In the meantime, the phosphorus was taken up in A/O/A operation mode. It is reasonably expected that the removal of both aniline and nutrients with high efficiency by the system might be an attractive alternative to aniline-containing wastewater treatment.

2. Methods

2.1. Sludge and media

Seed sludge inoculated into the SBRs was obtained from the secondary sedimentation tank of Shahu municipal wastewater treatment plant in Wuhan, Hubei Province, China. The initial COD loading rate fed in R1 and R2 was 0.9 g COD L^{-1} d^{-1}. The composition of initial synthetic wastewater for R1 was prepared with analytical-grade chemicals as follow (per liter): aniline 125 mg, NH_4Cl 50.2 mg, $NaNO_3$ 79.7 mg, KH_2PO_4 21.9 mg, $K_2HPO_4{\cdot}3H_2O$ 36.8 mg, CH_3COONa 192.3 mg and 1 ml of trace element solution. As for R2, the initial influent contained (per liter): CH_3COONa 576.9 mg, NH_4Cl 86.0 mg, $NaNO_3$ 136.6 mg, KH_2PO_4 21.9 mg, $K_2HPO_4{\cdot}3H_2O$ 36.8 mg and 1 ml of trace element solution. The trace element solution contained the following ingredients (per liter): $FeCl_3{\cdot}6H_2O$ 5.0 g, H_3BO_3 0.10 g, $CuSO_4{\cdot}5H_2O$ 0.10 g, KI 0.20 g, $MnCl_2{\cdot}4H_2O$ 0.50 g, $Na_2MoO_4{\cdot}2H_2O$ 0.20 g, $ZnSO_4{\cdot}7H_2O$ 0.30 g, $CoCl_2{\cdot}6H_2O$ 0.15 g, EDTA-2Na 10.0 g.

2.2. Reactor operation

Investigations were conducted in two parallel SBRs made of plexiglass. As shown in Fig. 1, the reactor consisted of two cylindrical columns with working volume of 9 L and the diameter/height (D/H) was 6.67. Influent synthetic wastewater was fed into the system through a pipe located at the top of the reactor by using a peristaltic pump and aeration was carried out through a porous stone diffuser at the bottom of the reactor. The sequential operation was automatically controlled by timers in the initial cycle of 6 h and the temperature of the reactors were maintained at 30 ± 2 °C by using water bath between the inner and outer columns. The initial operation was as follows: 5 min of feeding, 120 min of anaerobic agitation phase, 120 min of aerobic phase, 90 min of anoxic agitation phase, 20 min of settling time and 5 min of effluent withdrawal phase. For effluent stage, 4.5 L of the supernatant was discharged from the reactor by a diaphragm pump with 50% of volume exchange ratio. Thus, the hydraulic retention time (HRT) was 12 h and the dissolved oxygen (DO) in the anaerobic phase, aerobic phase and anoxic phase were controlled at about 0.05, 5.5, 0.1 mg L^{-1}, respectively. After the system was stable, the organic loading rate, DO in aerobic phase as well as HRT were varied in order to investigate the pollutants removal abilities of the two reactors with the variations in operation parameters. The operating conditions were shown in Table 1. The changes of process parameters of the two reactors are at the same time. Total nitrogen (TN) in influent was adjusted accordingly to remain the same ratio of carbon and nitrogen source throughout the whole operation. The samples were taken from reactors at 1 or 1.5 h intervals and residual concentration of aniline, COD, NH_4^+-N, TN and TP were tested. During the whole experiments, the concentrations of mixed liquor suspended solids (MLSS) in the two reactors were adjusted to 4200 ± 300 mg L^{-1}.

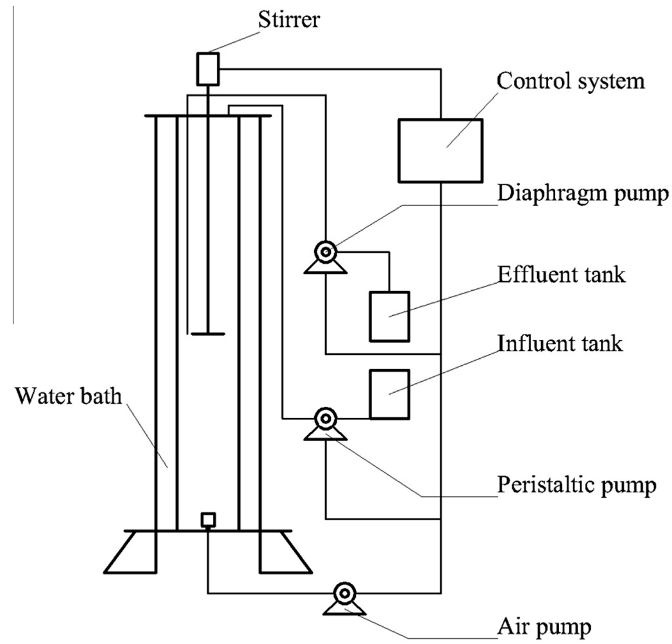

Fig. 1. Schematic diagram of the reactors.

Table 1
Operating condition of each stage in two SBRs.

Stages	Cycles	Days	COD loading rate (g COD L^{-1} d^{-1})	DO in aerobic phase (mg L^{-1})	HRT (h)
Stages 1	1–40	1–10	0.9	5.5 ± 0.5	12
Stages 2	41–80	11–20	1.5	5.5 ± 0.5	12
Stages 3	81–120	21–30	2.7	5.5 ± 0.5	12
Stages 4	121–160	31–40	3.9	5.5 ± 0.5	12
Stages 5	161–200	41–50	1.5	4.0 ± 0.5	12
Stages 6	201–240	51–60	1.5	5.5 ± 0.5	12
Stages 7	241–280	61–70	1.5	7.0 ± 0.5	12
Stages 8	281–340	71–80	1.5	5.5 ± 0.5	8
Stages 9	341–380	81–90	1.5	5.5 ± 0.5	12
Stages 10	381–410	91–100	1.5	5.5 ± 0.5	16