Reduction of excess sludge production in sequencing batch reactor through incorporation of chlorine dioxide oxidation

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ABSTRACT

In this study, chlorine dioxide (ClO_2) instead of chlorine (Cl_2) was proposed to minimize the formation of chlorine-based by-products and was incorporated into a sequencing batch reactor (SBR) for excess sludge reduction. The results showed that the sludge disintegrability of ClO_2 was excellent. The waste activated sludge at an initial concentration of 15 g MLSS/L was rapidly reduced by 36% using ClO_2 doses of 10 mg ClO_2/g dry sludge which was much lower than that obtained using Cl_2 based on similar sludge reduction efficiency. Maximum sludge disintegration was achieved at 10 mg ClO_2/g dry sludge for 40 min. ClO_2 oxidation can be successfully incorporated into a SBR for excess sludge reduction without significantly harming the bioreactor performance. The incorporation of ClO_2 oxidation resulted in a 58% reduction in excess sludge production, and the quality of the effluent was not significantly affected.

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1. Introduction

The wide use of an activated sludge process for wastewater treatment results in the production of a considerable amount of excess sludge, which must be safely disposed of due to its potential environmental risk. However, the disposal of these solid wastes is extremely costly, which may account for up to 60% of the total operating costs of a wastewater treatment plant [1]. The high cost is impractical, especially for municipal wastewater treatment plants that operate on a large scale. Therefore, excess sludge disposal represents an increasing challenge for wastewater treatment plants.

Several technologies, which involve mechanical, chemical, thermal and biological methods, have been explored for excess sludge reduction [2]. Among these technologies, chemical oxidation processes have shown great potential but high costs limit their application [3]. Biological methods present a relatively inexpensive alternative, however, sludge reduction through the manipulation of the ecosystem appears to be difficult in full-scale aerobic wastewater treatment processes.

An ideal method of solving excess sludge-associated problems is to reduce sludge production during wastewater treatment rather than during post-treatment of the sludge produced. Recently, some researchers have focused on combined process by incorporating chemical oxidation into activated sludge processes. Ozonation [4,5], chlorination [6], hydrogen peroxide oxidation [7] and fenton oxidation [8] have been successfully incorporated into the activated sludge process for excess sludge reduction. Chemical oxidation-assisted sludge reduction processes are more efficient for excess sludge reduction and have the advantages of easy control, stable performance and high operation flexibility.

Because it is a strong oxidant, ozone (O_3) has been widely applied in excess sludge reduction. A combined activated sludge process and ozone oxidation system has been successfully developed. In this combined system, the excess sludge production can be greatly reduced with the partial ozonation of the returned sludge from the activated sludge process [5,6]. A disadvantage of the ozonation-assisted sludge reduction process is its high operational cost because the generation and application of ozone is expensive.

Improving the cost effectiveness of such a sludge minimization requires the use of chemical oxidizers. Chlorine (Cl_2) was proposed to replace O_3 and was used in reducing excess sludge. Combining chlorination with an activated sludge process resulted in a 65% reduction in excess sludge production [6]. Although the

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Table 1
Wastewater treatment performance in the operation cycle of the SBR.

Parameters	Influent		Effluent		Removal rate
	Range	Average	Range	Average	
Total suspended solid (TSS, mg/L)	60–450	255	7.5–13.5	9.71	96%
Chemical oxygen demand (COD, mg/L)	100–500	300	20–30	26.5	91.2%
Biochemical oxygen demand (BOD$_5$, mg/L)	80–190	300	5.2–17.5	11.6	92%
Total nitrogen (TN, mg/L)	20–40	30	9–17.5	12.5	58%
Total phosphorus (TP, mg/L)	2–8	5	0.42–0.64	0.56	89%
Ammonium nitrogen (NH$_4^+$-N, mg/L)	5–30	17.5	1.0–2.5	2.1	88%
pH	6.8–7.8	7.3	6.8–7.8	7.3	–

chlorination-activated sludge process is more cost-effective than the ozonation-activated sludge system, the chlorination-generated potential harmful byproducts, including trihalomethanes (THMs), pose a serious challenge to the full-scale application of this technology.

As a novel neutral oxy-chlorine species, chlorine dioxide (ClO$_2$) has often been used as an alternative chemical oxidant for Cl$_2$ to minimize the formation of chlorine-based by-products [9,10]. ClO$_2$ is a more powerful oxidant than Cl$_2$, and ClO$_2$ accepts five electrons when reduced to a chloride ion. Based on its molecular weight and the number of transferred electrons, ClO$_2$ has approximately 263% more available chlorine, which is greater than 2.5 times the oxidizing capacity of the chlorine in Cl$_2$ [11]. Recent studies have focused on the use of ClO$_2$ to oxidatively eliminate undesired organic pollutants from surface water and wastewater [12–14]. Combining ClO$_2$ oxidation with an activated sludge process may is a promising technology for the minimization of excess sludge production.

To the best of our knowledge, there have been no reports related to ClO$_2$-assisted activated sludge reduction in the literature. In this study, we are the first to demonstrate that ClO$_2$ oxidation can be successfully incorporated into an activated sludge process to achieve excess sludge reduction. Excess sludge from a sequencing batch reactor (SBR) was subjected to various ClO$_2$ doses and the ClO$_2$ treated sludge liquor was then returned to the SBR system upon the completion of the ClO$_2$ treatment. To evaluate the feasibility of the proposed method, this study investigated the sludge disintegration ability of ClO$_2$, its sludge reduction efficiency and the effluent quality of the SBR system.

2. Materials and methods

2.1. Waste activated sludge cultivation

A SBR from the Nanhai municipal wastewater plant, which is located in Foshan, China was used to cultivate the waste activated sludge. The SBR, which had a working volume of 1260 L, was operated for two cycles per day, and the distribution of each 8 h cycle was as follows: an influent period (0.5 h), an aeration period (4 h), a precipitation period (1.5 h), a drainage period (0.5 h) and an idle period (1.5 h). The airflow rate was fixed at 700 L/h which maintains the DO at approximately 2–3 mg/L in the aeration phase. During steady operation, a portion of the mixed liquor was regularly discharged to maintain a mixed liquor suspended solids (MLSS) concentration of approximately 3500 mg/L and a sludge retention time (SRT) of 11.6 days so that the excess sludge was cultivated under sludge loading rate of 0.112 kg BOD$_5$/kg MLSS/day. The SBR was operated for more than 12 months and was controlled by a programmable logic controller (PLC). The composition of the domestic wastewater and wastewater treatment performance during the operation cycle of the SBR are shown in Table 1.

2.2. ClO$_2$ disintegration of waste activated sludge

The sludge disintegrability of ClO$_2$ was studied using beaker experiments. We subjected 100 mL of waste activated sludge at a concentration of 15 g/LMLSS to varying doses of ClO$_2$ from 1 to 20 mg ClO$_2$/g dry sludge to investigate the effect of the ClO$_2$ dose on sludge disintegration and to determine the optimal ClO$_2$ dose. Subsequently, sludge disintegration by ClO$_2$ was evaluated at the previously determined optimal ClO$_2$ dose, and the MLSS, soluble chemical oxygen demand (SCOD), nitrogen and phosphorus were monitored. Highly pure ClO$_2$ (95%) was supplied by a ClO$_2$ generator (HYCB-50, China). ClO$_2$ was generated using the hypochlorite reaction with hydrochloric acid according to Eq. (1).

$$5NaClO_2 + 4HCl \rightarrow 4ClO_2 + 5NaCl + 2H_2O \tag{1}$$

All of the experiments were conducted at least in duplicate at room temperature (25 ± 2 °C) and the average value was reported for all data.

2.3. ClO$_2$-activated sludge process combined system

Fig. 1 presents a schematic representation of the ClO$_2$-activated sludge process combined system. The previously described SBR was combined with a separated ClO$_2$ oxidation reactor in which highly pure ClO$_2$ (95%) was supplied by a ClO$_2$ generator. The ClO$_2$ oxidation reactor was a plexiglass cylinder with a working volume of 5 L (150 mm in diameter and 355 mm in height). A coarse bubble diffuser was placed at the bottom of the reactor to disperse pure ClO$_2$ gas, and a magnetic stirrer was used to facilitate the reaction between the sludge and ClO$_2$. ClO$_2$ doses were controlled using a manual valve. During operation, 50% of the excess sludge in the SBR was pumped into the ClO$_2$ oxidation reactor, in which the sludge was disintegrated. The disintegrated sludge was then returned to the SBR for further biological treatment and the effluent quality was monitored daily. An identical SBR that did not incorporate ClO$_2$ oxidation was used as control. Both SBRs were fed with the same domestic wastewater (Table 1) and operated at room temperature (25 ± 2 °C) for 12 months.

2.4. Analytical methods

SCOD, MLSS, TN, NH$_4^+$-N, and TP and were measured according to standard methods [15]. For the SCOD measurement, all samples were filtered through a 0.22 um-pore-size syringe filter unit.

Protein content was determined using Lowry's method and polysaccharide content was analyzed using the anthrone method [16].

Sludge morphologies before and after ClO$_2$ treatment were determined using a scanning electron microscope (SEM) (XL-30, Philips, Holland). Before observation, a sample was collected and fixed overnight using paraformaldehyde and glutaraldehyde in a buffer solution (0.1 M cacodylate, pH 7.5, at 4 °C), followed by
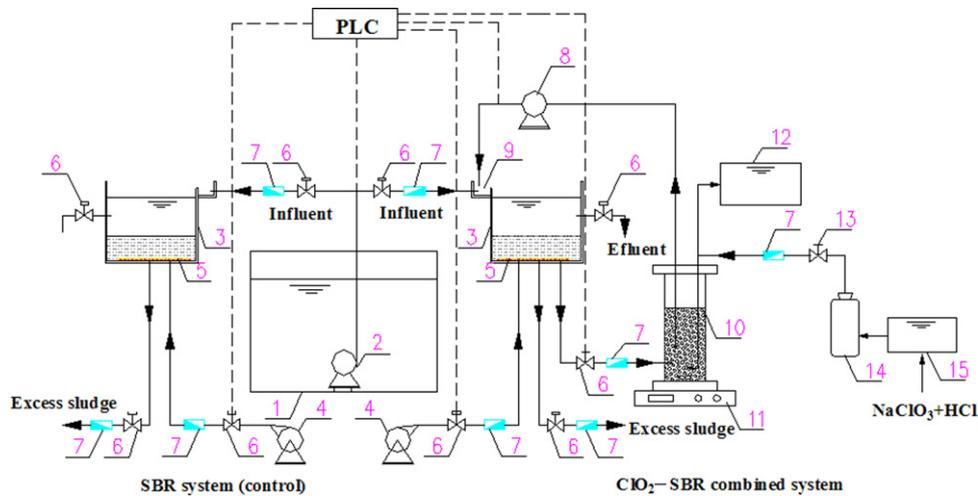
Fig. 1. Schematic illustration of ClO$_2$-activated sludge process combined system: (1) wastewater storage tank; (2) submersible pump; (3) SBR system; (4) air compressor; (5) bubble diffuser; (6) electromagnetic valve; (7) flow meter; (8) disintegrated sludge circulator pump; (9) electromagnetic valve; (10) ClO$_2$ oxidation reactor (11) magnetic stirrer; (12) ClO$_2$ absorption tank (NaOH); (13) manual valve; (14) ClO$_2$ storage tank (purity: 95%); (15) ClO$_2$ generator.

washing and dehydration in water/ethanol. The samples were then coated with Au/Pt before SEM observation

3. Results and discussion

3.1. Sludge disintegrability of the ClO$_2$

In general, the disintegration of the sludge floc and the cell results in the release of organic matter, which can be quantified using parameters such as MLSS content, SCOD, protein, polysaccharide, nitrogen and phosphorus in the mixed liquid containing the sludge. To test the ClO$_2$ disintegrability of the wasted activated sludge (15 g/L of MLSS), a series of experiments were performed at different ClO$_2$ doses and with a reaction duration of 60 min. The results suggested that the sludge disintegrability of the ClO$_2$ was excellent. As shown in Fig. 2, increasing ClO$_2$ doses from 1 to 10 mg ClO$_2$/g dry sludge resulted in a sharp reduction in the MLSS content which was accompanied by a significant increase in the SCOD. At ClO$_2$ doses of 10 mg ClO$_2$/g dry sludge, a decrease of 5.4 g/L of MLSS and an increase of 92.5 mg/L of SCOD was observed. However, a further increase in the ClO$_2$ doses did not result in a further reduction in the MLSS. Similar changes were observed for the concentration of protein and polysaccharide in solution which also indirectly

indicated the disintegration of the sludge due to cell content leakage (Fig. 3). However, this phenomenon was not observed another study that used ozone for sludge disintegration which could be attributed to the fast oxidation of protein and polysaccharide by ozone [17]. Because it is an inferior oxidant to ozone, the ClO$_2$ used in this study was not powerful enough to quickly oxidize the proteins and polysaccharides. The observed results suggested that ClO$_2$ doses of 10 mg ClO$_2$/g dry sludge are desirable. Compared with a previous study that used Cl$_2$ to reduce sludge, our ClO$_2$ dose is much lower than that of the previous study, which was 332 mg Cl$_2$/g MLSS based on similar sludge reduction efficiency [6]. Therefore, 10 mg ClO$_2$/g dry sludge was determined to be the optimal dose of ClO$_2$ and was used for further studies.

To achieve high sludge reduction efficiency and simultaneously reduce costs, the duration of the ClO$_2$ oxidation process should be determined. Fig. 4 displays the degree of sludge reduction by ClO$_2$ oxidation as a function of reaction time. The duration of 60 min for the ClO$_2$ oxidation process was divided into two stages. A sharp reduction in MLSS, significant increase in SCOD was observed until the reaction time reached 40 min. However, a longer reaction time only resulted in a slight decrease in MLSS. Based on the observed results, we determined an optimal reaction time of 40 min.

The concentration of nitrogen and phosphorus were also measured to confirm the effect of ClO$_2$ oxidation on sludge reduction.
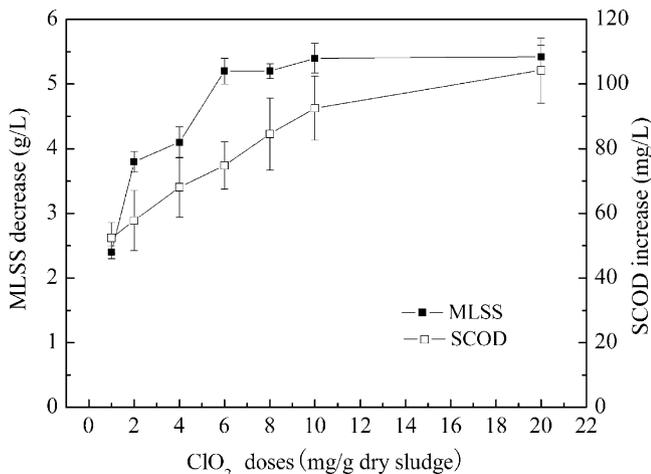
Fig. 2. Degree of MLSS reduction and SCOD release at increasing ClO$_2$ doses (Errors bars present the standard deviation of the measurements).
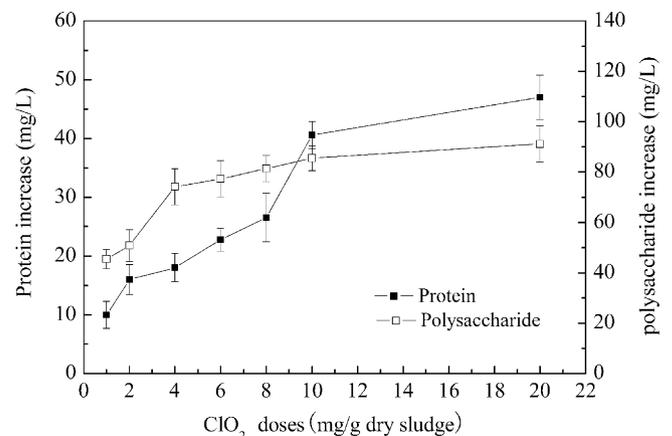
Fig. 3. Degree of protein and polysaccharide increase in solution at increasing ClO$_2$ doses (Errors bars present the standard deviation of the measurements).
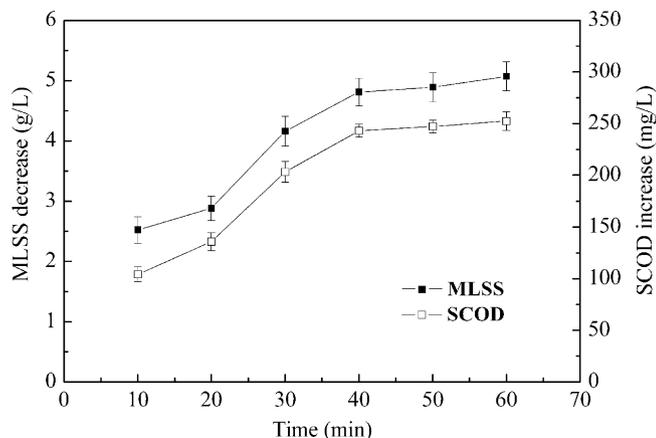
Fig. 4. Degree of MLSS reduction and SCOD release with respect to the reaction time (ClO$_2$ doses: 10 mg/g dry sludge, errors bars present the standard deviation of the measurements).
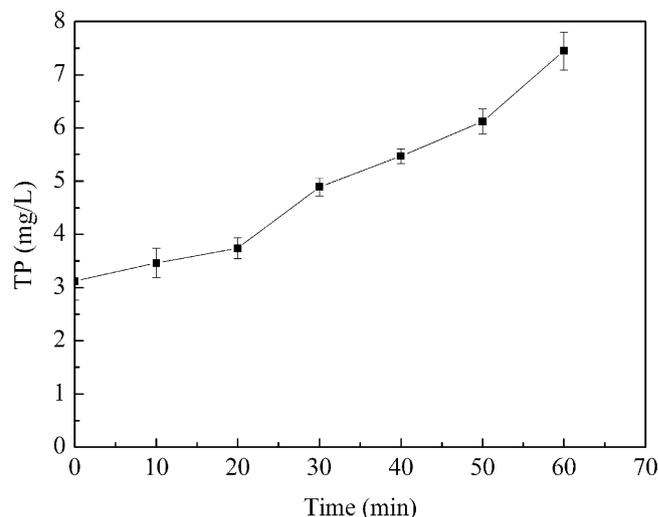
Fig. 5. Variations of TN and N-NH4 in the mixed liquid with respect to the reaction time (ClO$_2$ doses: 10 mg ClO$_2$/g dry sludge, errors bars present the standard deviation of the measurements).

As shown in Fig. 5, a significant rise in the concentrations of NH$_3$-N and TN was clearly visible during the ClO$_2$-assisted sludge disintegration process. The variation of the concentration of TN and NH$_3$-N was similar to that of the MLSS reduction. ClO$_2$ oxidation resulted in a sharp increase in both the concentrations of TN and NH$_3$-N during a reaction time of less than 40 min. We observed a 264.43% increase in TN and 70.24% increase in NH$_3$-N. By increasing reaction time to 60 min, the concentration of TN remained stable, whereas the concentration of NH$_3$-N began to decrease. Compared to nitrogen, TP concentration continuously increased even when the duration of the reaction reached 60 min (Fig. 6).

The SEM images of the untreated and treated sludge (10 mg ClO$_2$/g dry sludge) are shown in Fig. 7. Fig. 7A shows that the floc of the untreated sludge was fully compacted, however, in the case of the treated samples (Fig. 7B), the floc became intumescent and loose. The SEM images indicated that the sludge floc was partly disintegrated. Cell wall breakage was contributed to the increase of SCOD, proteins, polysaccharides, nitrogen and phosphorus.

3.2. SBR performance after incorporation of ClO$_2$ oxidation

3.2.1. Excess sludge production

To investigate the effect of ClO$_2$ oxidation on the performance of the activated sludge process, ClO$_2$ oxidation was incorporated into an SBR system at selected conditions. During the stable opera-

Fig. 6. Variations of TP in the mixed liquid with respect to the reaction time (ClO$_2$ doses: 10 mg ClO$_2$/g dry sludge, errors bars present the standard deviation of the measurements).
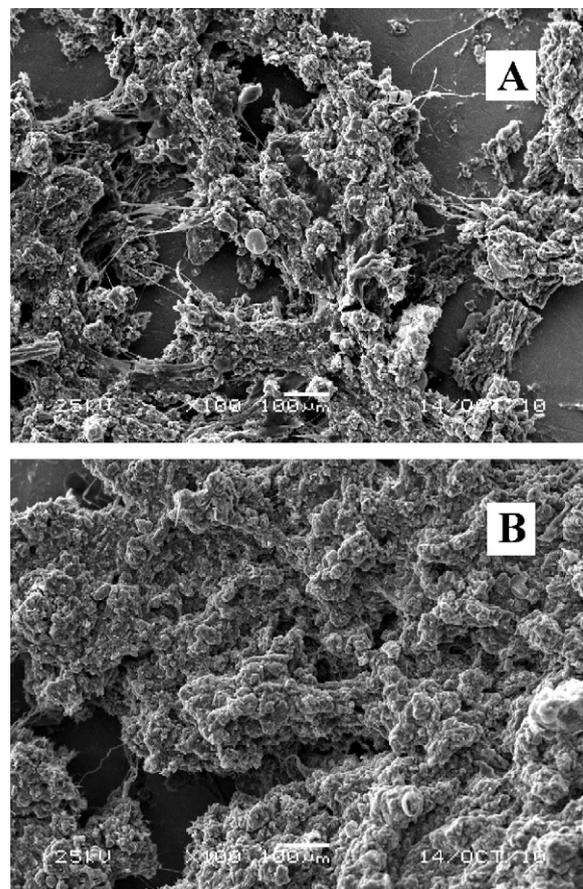
Fig. 7. SEM images of sludge before (A) and after treated (B) by the ClO$_2$ oxidation (10 mg ClO$_2$/L dry sludge).

tion of the testing system, 50% of the excess sludge was treated at a ClO$_2$ dose of 10 mg ClO$_2$/g dry sludge for 40 min in the ClO$_2$ oxidation reactor, and then it was returned slowly. Fig. 8 shows that the incorporation of ClO$_2$ oxidation significantly reduced excess sludge production in the SBR system. The decrease in sludge production appeared very quickly after the incorporation of the ClO$_2$ oxidation. The daily sludge production rate over 22 days was maintained
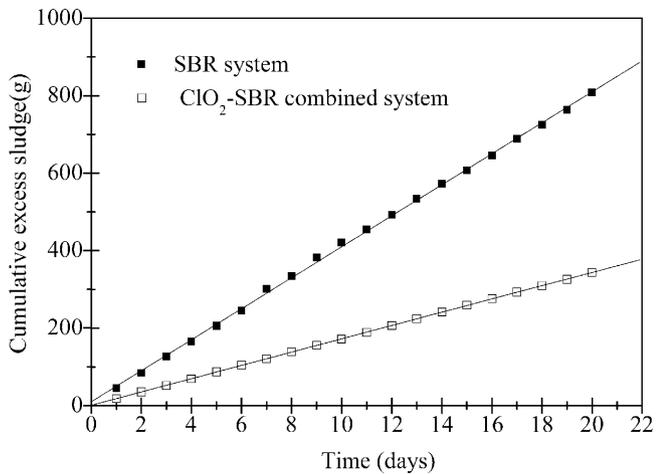
Fig. 8. Sludge production rates in the SBR system (the reference system) and ClO_2–SBR combined system (ClO_2: 10 mg/g dry sludge).

at approximately 17 g MLSS/day, which was an approximate 58% reduction of efficiency compared with that of the control SBR system (40 g MLSS/day). The sludge reduction efficiency obtained here was significant, although it was slightly lower than that which was previously reported using chlorination, which was 65% at a Cl_2 dose of 66 mg Cl_2/g MLSS [6]. Using a smaller ClO_2 dose coupled with higher sludge reduction efficiency suggests that the incorporation of ClO_2 oxidation into an activated sludge process is very effective and is able to replace the Cl_2 and ozonation during excess sludge reduction.

3.2.2. Effluent quality

To investigate ClO_2 treatment on the effluent quality of SBR, SS, SCOD, NH3-N, TN and TP in the effluent were monitored for 22 days. Fig. 9 presents the variations of SS, SCOD, TN and TP plotted with respect to the operating time in both the control SBR and the ClO_2–SBR combined system.

Fig. 9A shows that the incorporation of sludge ClO_2 oxidation resulted in a slight increase in the effluent SS concentration during the SBR process. The average SS concentration in the effluent of the ClO_2–SBR combined system increased by 30% compared with the control SBR system (without sludge ClO_2 oxidation). The increased SS could be attributed to the cell fragments or inorganic larger particles that occurred due to sludge disintegration. Although the incorporation of ClO_2 oxidation resulted in an increase in the SS of the effluent, sludge settleability was not deteriorated, the SS could be easily removed through settling. It was notable that, the total removal efficiency of SS was not affected by the oxidation of ClO_2. Similar SS removal efficiencies of 96.2% and 97.3% were observed in the ClO_2 oxidation–SBR combined system and the control SBR, respectively. Moreover, the average SS concentration in the effluent met the first-class B discharge standards for a municipal wastewater treatment plant in China (<20 mg/L).

As is shown in Fig. 9B, C and D, the SCOD, NH_3-N and TN concentrations in effluent were not affected by sludge ClO_2 oxidation. They were maintained at a satisfactory level, which was less than 20 mg/L for SCOD, 2.5 mg/L for NH_3-N and 17.5 mg/L for TN. Due to ClO_2 oxidation, the sludge was disintegrated and the cell contents were leached into the supernatant, which contributes to the increase in SCOD and nutrients in the effluent. In this study, both of these were removed using coinstantaneous biological process, which suggests that the predominance of the ClO_2–SBR combined sys-
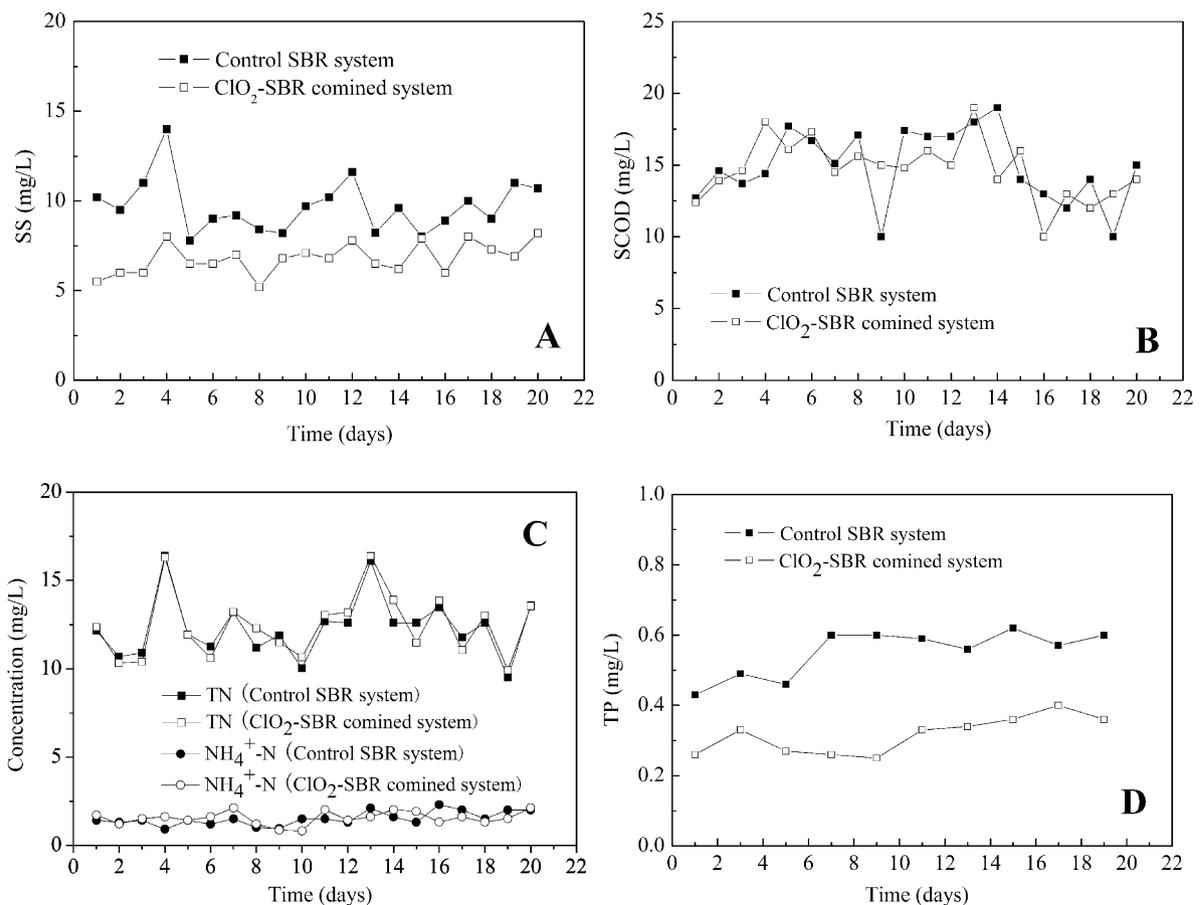
Fig. 9. Variations of the SS (A), SCOD (B), TN (C) and TP (D) in the effluent of the ClO_2–SBR combined system, with single SBR system used as control.

tem was dominant over the control SBR system. Previous research shown that general heterotrophic bacteria and nitrifying bacteria were deactivated at an ozone dose of 0.05 g O_3/g TSS [18,19]. In this study, the further removal of SCOD and nitrogen indicated that the activity of these bacteria was least affected by the ClO_2 oxidation at a ClO_2 dose of 10 mg ClO_2/g dry sludge. In addition, a portion of SCOD from the ClO_2 oxidated sludge may have been used as a carbon source for denitrification, which could have partially contributed to the removal of SCOD from the effluent [20]. The ClO_2 oxidation-activated sludge process also has advantages over a chlorination-activated sludge system, in which poor sludge settleability and a significant increase of soluble COD in the effluent has been observed [6].

However, the TP concentration in the effluent of the ClO_2–SBR combined system was observed to be 43% higher than that of the control SBR system (Fig. 9D). The phosphorus release was unavoidable because biological phosphorus removal mainly depends on the bioabsorption and bioaccumulation. Similar phenomena have also been observed in ozonation-activated sludge systems [21,22]. It is noteworthy that the average TP concentration in the effluent of the ClO_2–SBR combined system was not as high (0.55 mg/L), which meets the first-class B discharge standards for a municipal wastewater treatment plant in China (1 mg/L) [23].

Overall, the ClO_2 oxidation–SBR combined system satisfactorily met the currently existing discharge standards for a municipal wastewater treatment plant in China.

4. Conclusions

First, ClO_2 oxidation for the disintegration of excess sludge generated in the activated sludge process was tested and incorporated into SBR for excess sludge reduction. We drew the following conclusions:

(1) The sludge disintegrability of the ClO_2 was excellent. The waste activated sludge at an initial concentration of 15 g MLSS/L was rapidly reduced by 36% at ClO_2 doses of 10 mg ClO_2/g dry sludge, which was much lower than that found by using Cl_2 based on similar sludge reduction efficiency.
(2) For 15 g MLSS/L of excess sludge, maximal disintegration was achieved at 10 mg ClO_2/g dry sludge for 40 min.
(3) ClO_2 oxidation was successfully incorporated into the SBR process for excess sludge reduction without significantly lowering the bioreactor performance. Excess sludge production was reduced by 58% after incorporating ClO_2 oxidation and the water quality of the effluent reached a satisfactory level.

Acknowledgment

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