A pilot-scale test on the treatment of biological pretreated leachate by the synergy of ozonation-biological treatment-catalytic ozonation

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ABSTRACT

In this pilot-scale test, the ozonation-biological treatment-catalytic ozonation system was performed to treat complex organics and highly-concentrated total nitrogen (TN) in biological pretreated incineration leachate. The test results showed that the ratio of five-day biochemical oxygen demand (BOD₅) / chemical oxygen demand (COD) increased from 0.059 to 0.237, which indicated that the concentration of biodegradable COD (COD_{bio}) increased by ozonation pre-treatment process. In addition, the TN removal mainly occurred in anaerobic zone due to direct denitrification by the activated bacteria, which were domesticated through different influent ratio. Moreover, it was necessary to add catalytic ozonation process to reach higher direct effluent discharge criteria. After 60 days repeated debugging, the removal rate of COD and TN reached 88.5% and 98.2%, respectively. Finally, the total cost of this system was ¥ 6.65 /m³ (\$ 0.95 /m³), which was acceptable for the treatment of biological pretreated leachate. This pilot-scale test could provide some guiding information for the treatment of leachate containing highly-concentrated TN with low COD_{bio}/N by the composite system.

Keywords: COD removal, Composite system, Incineration leachate, TN removal

1. Introduction

Incineration has been an effective technology of municipal solid waste (MSW) disposal for its advantages in reducing approximately 90% of the volume and 75% of the mass from MSW, energy production and saving space [1, 2]. The fresh MSW in general needs to be held for 3-7 days before incineration to remove its high moisture content, but unfortunately a considerable amount of fresh leachate can be produced during this period [3]. Leachate will be generated by liquid from solid wastes, which has high contaminant concentrations [4]. Thus, incineration leachate is a complex wastewater which contains a large amount of biodegradable organic, refractory organics, heavy metals, ammonia and many other toxic matters [5, 6]. In conventional engineering applications, biological treatment followed by membrane separation technique such as membrane

bio-reactor (MBR), reverse osmosis (RO), nanofiltration (NF) and ultrafiltration (UF), they had been widely applied in the treatment of leachate [7]. Since the majority of easily biodegradable organics in leachate are removed through the above-mentioned combined process, most of the remaining matters are made of humic substances with aromaticity and macromolecular weight which lead to low biodegradability (BOD₅/COD < 0.1). At present, the main treatment technology of biological pretreated and membrane separated leachate are the recirculation and advanced oxidation processes (AOPs) [8, 9]. However, the recirculation of leachate may inhibit the microbial activity and affect the performance of membrane separation process due to the high concentrated organics and heavy metals [10, 11]. And if lots of organic matters and heavy metals in incineration leachate are not treated properly, they would pollute surface water, groundwater, and change soil quality. Therefore,

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Received June 21, 2020 Accepted August 15, 2020

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it is urgent to explore appropriate technologies to treat incineration leachate [12].

According to the previous studies, some methods have been used to treat leachate, such as electrochemical oxidation [13, 14], Fenton process [15-17], ozone-based advanced oxidation process [18, 19], etc. Electrochemical oxidation although having versatility, high efficiency and environmental compatibility in handling leachate, have shortcomings of being energy-intensive process and high operating costs [20]. Fenton process could effectively degrade organics via powerful oxidizing agents. However, it also has shortcomings such as the generation of iron sludge and narrow pH range between pH 2-3 [18, 20]. Compared with the above-mentioned methods for the leachate, ozonation has wide engineering application in wastewater because of its excellent oxidation capacity and no secondary pollution property [21]. Ozone molecules, ozone self-decomposing hydroxyl radicals ($\bullet\text{OH}$) or a combination of O_3 and $\bullet\text{OH}$ can react with refractory organics [22, 23]. Overall 1.5 mol of O_3 could produce 1 mol of $\bullet\text{OH}$ [24]. According to standard oxidation potentials, $\bullet\text{OH}$ ($E^0 = 2.8 \text{ v}$) with excellent reaction rate constants of $10^6 - 10^9 \text{ M}^{-1} \text{ S}^{-1}$ is stronger oxidant than molecular ozone ($E^0 = 2.08 \text{ v}$), which has the ability to oxidize the complex organics, especially the humic substances [25, 26]. Ozonation process alters the molecular structure of refractory compounds and oxidize them to more biodegradable compounds used as carbon source for the denitrification in subsequent biological treatment [18, 20]. Therefore, ozonation is an effective approach as pre-treatment of leachate to improve biodegradability. Catalyst can be an add-on to accelerate reaction in ozonation. Catalytic ozonation can effectively remove organic compounds since catalysts promote surface reactions between the adsorbed ozone and pollutants. To meet stricter effluent discharge criteria, catalytic ozonation further degrade organic pollutants as post-treatment. H. Wang [19] showed that micro-ozonation could achieve 76.0% COD and 69.9% TOC (Total Organic Carbon) removal for the further treatment of refractory substances in leachate under optimal conditions.

It has been observed that AOPs combined with biological treatment for the leachate could reach direct effluent discharge criteria. The biological treatments are the common technique for removing nitrogen owing to the simplicity and cost-effectiveness [27]. The existing nitrogen removal processes are mainly including conventional nitrification-denitrification, nitrification-denitrification and anammox process, etc [28]. F.N. Ahmed [29] found that denitrification-partial nitrification-anammox process could utilize COD_{bio} in wastewater to achieve the NO_3^- -N removal, and was also beneficial for the growth of autotrophic bacteria. Z. Wang [30] reduced the NO_3^- -N through partial denitrification-anammox process. However, there are few studies about the engineering application of ozonation-biological treatment-catalytic ozonation for the biological pretreated and membrane separated leachate.

The typical characteristics of the biological pretreated and membrane separated leachate from this test are macromolecular (organic matters), highly-concentrated (total nitrogen) and refractory (low biodegradability). Therefore, this pilot-scale test devoted to degrading macromolecular organic matters and removing highly-concentrated TN by the synergy of ozonation-biological treatment-catalytic ozonation. Especially the main purpose of the test is to remove TN due to the low $\text{COD}_{\text{bio}}/\text{N}$ ratio in leachate, which

is the most challenging part of this pilot-scale test. In this work, ozonation was selected as the pre-treatment process to decompose macromolecular organic contaminants into biodegradable one and increase the $\text{COD}_{\text{bio}}/\text{N}$ ratio, which was beneficial for the subsequent nitrogen removal in the biological process. Then in the biological process, nitrogen was removed by activated bacteria using organic carbon as electron donor and nitrate as electron acceptor. At the end, to further optimize the effluent, catalytic ozonation was used to deeply remove organic pollutants. The pilot-scale test will provide useful information for full-scale practical implementation.

2. Material and Methods

2.1. The Raw Water

The raw water used in this test was got from an MSW incineration power plant in the Everbright Environmental Energy (Nanjing) Co., Ltd., China. The water samples were collected after biological pre-treatment and ultrafiltration separated process. The characteristics of the raw water are shown in table 1. Values are given as an average.

Table 1. Initial Characteristics of Wastewater

Parameters	Values	Parameters	Values
COD (mg/L)	389-513	Color	Dark brown
NH_4^+ -N (mg/L)	16.5-22.3	Smell	light
NO_3^- -N (mg/L)	850-960	B/C ratio	0.059
TN (mg/L)	1070-1200	pH	7-8

2.2. Materials

Activated bacteria (a milky liquid, cfu > ten billion per milliliter) and composite carbon source (a dark brown liquid, smell aromatic) were provided by Shenzhen Changlong Technology Co., Ltd., China.

2.3. Experimental Procedures

Based on the property of raw water, the pilot-scale setup (Fig. 1) including ozone and biological system was designed, and the related research results were tested in laboratory. The ozone system was consisted of ozone generator (1 kg O_3/h), ozonation reaction column (with a diameter of 0.5 m and a height of 2 m), catalytic ozonation reaction chamber (2 m [L] $\times$ 0.5 m [W] $\times$ 2 m [H]), ozone exhaust destructor, and ozone analyzer (read ozone concentration in real time). The biological system was composed of back-washing pump, reflux pump, agitator and aerator, containing anaerobic zone (0.3 m [L] $\times$ 1.4 m [W] $\times$ 2.1 m [H]), anoxic zone (0.5 m [L] $\times$ 0.5 m [W] $\times$ 2 m [H]), aerobic zone (0.4 m [L] $\times$ 0.5 m [W] $\times$ 2 m [H]), and ceramic membrane separation zone (1 m [L] $\times$ 0.5 m [W] $\times$ 2 m [H]). Ozone was generated from oxygen conversion in ozone generator and simultaneously entered to the ozonation reaction column. The raw water was pre-treated by ozonation and then entered to the biological treatment system. After the anaerobic, the anoxic and the aerobic treatment, the sewage returned or entered to ceramic membrane pool. Finally, in order to attain higher direct effluent discharge criteria, the membrane effluent would be treated

Fig. 1. Experimental apparatus for pilot-scale test.

by catalytic ozonation. In the biochemical system, three backflow paths were set up. Reflux 1: from the ceramic membrane pool to aerobic pool 1 and anoxic pool, the sludge with influent flow to the ceramic membrane pool was returned back to the front end, which at the same time diluted the influent, increased the sludge concentration and the processing capacity of aerobic pool 1 and anoxic pool. Reflux 2: from the aerobic pool 3 to anaerobic pool 2, the process improved the denitrification. Reflux 3: from anaerobic pool 3 to anaerobic pool 1, the anaerobic sludge that came with the influent to anaerobic pool 3 would return to the front end, and the anaerobic wastewater would be diluted, simultaneously the sludge concentration and the processing capacity of anaerobic pool 1 would be enhanced.

2.4. Inoculated Sludge

The activated sludge used in the test was got from in the aerobic pool of MSW incineration power plant. The initial concentration of mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solid (MLVSS) and sludge volume index at 30 min (SVI_{30}) of activated sludge were 5.0 mg/L, 3.5 mg/L, 75%, respectively. The method of inoculated sludge of different zones in the biological system was as follows: the sludge and leachate were fully mixed at a volume ratio of 3:20, and then was poured into biological zones. According to the $COD_{bio} : N = 6 : 1$, homemade synthetic carbon source was added to increase electron donor in the anaerobic zone for the inoculation of activated bacteria provided by Shenzhen Changlong Technology Co., Ltd., China. The activated bacteria were cultured to remove TN in anaerobic zone. The regular dosage of activated bacteria is 10 g/m³, and 3 times of the regular dosage should be required for the first 3 days. Then add activated bacteria continuously for 7 days according to the regular dosage. The aerobic bacteria were cultured under complete aeration in the aerobic zones. The concentration of DO in the aerobic zone was controlled to be 2 mg/L at room temperature. pH of system was stable at 7-8. In order to adapt highly-concentrated TN caused by leachate concentrates, the biological system was

performed by three stages. The system took sequential batch influent, the proportion of feed water was 15%, 25% and 41%, respectively.

2.5. Analytical Methods

The COD, NH_4^+-N , NO_3^--N , TN, MLSS, MLVSS and SV_{30} were measured according to the standard methods [31]. The pH was measured using a pH meter (pHBJ-261L, Shanghai Scientific Instrument Co., Ltd., china). The DO was measured using a DO meter (BDO-7500, BELL Analytical Instrument (Dalian) Co., Ltd., china).

3. Results and Discussion in Different Stages

3.1. Ozonation Stage

As a promising method, Ozonation is suitable for leachate pre-treatment, which could improve the biodegradability and reduce wastewater toxicity. In this stage, the macromolecular organics are broken into easily biodegradable smaller ones by ozonation pre-treatment to increase the concentration of COD_{bio} as carbon source for subsequent denitrification in wastewater [32]. As shown in Fig. 2 (a), this process improved the biodegradability, with B/C average rising from 0.059 to 0.237, laying a foundation for subsequent biological system [33, 34]. The parameters of ozonation are mainly decided by adjusting the influent flow, power of ozone generator and gas flow, changing the water retention time and ozone dosage.

The influent flow of ozonation pre-treatment was 0.2 m³/h, the total gas flow was controlled to 0.5 m³/h, the gas flow of pre-treatment was designed to 0.3 m³/h, the power of ozone generator was adjusted to obtain different ozone concentration and the corresponding ozone dosage would change. After half a month of repeated debugging, the test results are shown in Fig. 2 (b). The results demonstrated that the average removal rate of COD gradually increased, after increasing to a certain degree then would not obviously increase.

In preliminary experiment, with the increasement of ozone dosage, the ability of directly oxidizing organic pollutants by ozone molecule was increased. Meanwhile the $\cdot\text{OH}$ (indirect oxidation) produced from the decomposition of ozone was also increased. This observation was similar to some report [35-37] that the degradation efficiency of organics increases with increasing ozone concentration. The ozone dosage increased from 120 mg/L to 132 mg/L, but the COD removal rate was not increased obviously due to the saturation of the aqueous ozone [38, 39]. In saturated stage, it was at equilibrium between the consumption and supply, the increasement of ozone concentration was redundant. It is worth noting that Barbara Kasprzyk-Hordern *et al.* [40] found that the degradation ability of organic contaminants would reduce for the reaction between the excessive amount of ozone and free radicals. Thus, the experiment selected 120 mg/L as the ozone dosage. In ozonation stage, the COD removal rate was no longer increased at about 38.8%, which was similar to the research [41]. The reason for the low efficiency was that the small decrease of COD, which was affected by the organic contaminant mineralization. And it is obvious that complete oxidation for the raw water containing highly-concentrated NO_3^- -N in this pilot-scale test is inappropriate, but partial oxidation destroying original macromolecular organic structure is beneficial by reducing toxicity and enhancing biodegradable organics as carbon source in subsequent biological treatment [37]. In the ozonation pre-treated process, when the ozone dosage was 120 mg/L, the COD concentration change in the influent and effluent is shown in Fig. 2 (c), the average COD of influent and effluent were 450 mg/L, 279 mg/L, respectively.

The average changes of NH_4^+ -N, NO_3^- -N and TN during ozonation

pre-treated process are shown in Fig. 3. After the ozonation process, the results showed increasing NO_3^- -N, slightly decreasing NH_4^+ -N and almost unchanged TN. In pre-treated process, a small amount of ammonia nitrogen could be oxidized to the nitrate nitrogen, and the nitrate nitrogen in ozone environment was mainly from the oxidation of the nitrite nitrogen, so the nitrate nitrogen increased obviously; In the case of strong oxidation, the radicals produced by ozonation potentially resisted ammonification from organic nitrogen, and a small amount of ammonia nitrogen may be oxidized to nitric nitrogen, the combination of both would reduce the concentration of ammonia nitrogen, therefore the trend of ammonia nitrogen was lightly decreasing [42]. The ozonation process couldn't remove nitrogen, which led to almost unchanged TN.

3.2. Biological Stage

Aiming to treat the highly-concentrated NO_3^- -N wastewater, direct denitrification is an economical method [30], so the experiment focused on controlling denitrification conditions and guaranteed sludge reflux in time among the zones. In this stage, the influent was fed into anaerobic zone to remove nitrogen by direct denitrification; the organic nitrogen in wastewater would be converted into NH_4^+ -N by amination in the anoxic zone, and then were removed by nitrification in aerobic zone; the remaining organic pollutants would degrade in aerobic zone.

The system inoculated bacteria by three different influent proportion of 15%, 25% and 41% [29, 43]. After three different influent ratios debugging, yellowish-brown sludge and larger flocs were observed, the sedimentation performance was better than before.

Fig. 2. Organics change in ozonation process: (a) B/C change, (b) COD removal rate, (c) COD change.

Fig. 3. The changes of nitrogen in ozonation process: (a) NH_4^+ -N, (b) NO_3^- -N, (c) TN.

Fig. 4. The change of (COD and TN) and (COD and TN) removal in the biological process by different influent ratio: (a) COD change, (b) COD removal change, (c) TN change, (d) TN removal change.

During the debugging, the influent ratio of 15%, 24% and 41% were tested, respectively. And the results were almost the same after operating with 24 h, 36 h and 56 h, respectively. The changes of COD and COD removal are shown in Fig. 4 (a) and (b). In Fig. 4 (a), the initial COD values are high because of the external carbon source. However, because the homemade carbon source was easily decomposed later, the COD could be controlled (less than 200 mg/L). At the stage of 15% influent ratio, the system took 24 h to achieve the expected result (COD was less than 200 mg/L). According to this method, when the system influent ratio was 25% and 41%, it would take 38.4 h and 65.6 h to obtain the similar result (COD was less than 200 mg/L). Surprisingly, the actual time to obtain the similar result was less than the theoretical time, and it just took 36 h and 56 h to reach the result when the influent ratios were 25% and 41%, respectively. This was because the low concentration of MLSS in the initial domesticated stage [43]. With the increase of influent ratio, the whole system gradually became mature and tended to be steady. After debugging, the sludge activity reached its optimum state (MLSS: 8.62 g/L, MLVSS: 6.87 g/L, SV_{30} : 55%), which had enough microorganisms to degrade organic contaminants. At the stable stage, the effluent COD average concentration was about 183 mg/L, the COD average removal obtained 81%.

As shown in Fig. 4 (c) and (d), with the influent ratio of 15%, 24% and 41%, the TN concentration obtains the similar results (TN: 15 - 20 mg/L) after operating the system by 24 h, 36 h and

56 h, respectively [44]. The removal of TN was correlated with the denitrification efficiency. At the initial debugging, the most prominent feature was the insufficiency of biodegradable organic matter. The biodegradable organic matters would be increased with the increase of influent ratio, which added denitrification electron donor [43]. With the increase of denitrification efficiency, the system would take less time to achieve the anticipated result (TN was less than 20 mg/L). The result was consistent with the report that the effluent TN was less than 20 mg/L by partial denitrification-anammox process [30].

Because the debugging was in accordance with the different influent ratio, the ammonia oxidation, nitrification and denitrification had achieved good results in the biological system. Therefore, according to the above method, the influent flow of the biological system with 1120 mg/L TN was designed to be 0.2 m³/h. The whole biological system could run well by 100% influent ratio, the average COD of the effluent is remained within 200 mg/L, and TN is about 15 ~ 20 mg/L, which indicate the method is practical.

3.3. Catalytic Ozonation Stage

Some literatures have been shown that the heterogeneous catalytic ozonation is a promising process for the degradation of refractory organic matters in post-treatment [45-47]. Therefore, the system selected a Fe-Cu/ γ -Al₂O₃ (with a diameter of 6 - 8 mm, Brunner-Emmet-Teller (BET) surface area was about 164.2 m²/g) composite

Fig. 5. The COD effluent concentration and the removal rate in catalytic ozonation process.

as catalyst to enhance removal efficiency. In this process, the gas flow was assigned to 0.2 m³/h and the ozone dosage was 80 mg/L, which was consistent with ozonation debugging. As shown in Fig. 5, the effluent COD is controlled within 50 mg/L. With the addition of 50 kg/m³ catalyst (the amount of catalyst was calculated by the result of laboratory test), 75.2% COD was removed. The ozone dosage was affected by the addition of the catalyst: when the pre-oxidation ozone dosage was 120 mg/L, the average COD removal rate was 38.8%; however, the COD removal rate reached to 75.2% when catalytic oxidation ozone dosage was 80 mg/L. This was because ozone and organic pollutants could be adsorbed on the catalyst's surface, and then generated the HO₃⁻ and •OH to degrade contaminants. As a result, the removal rate was doubled by using less ozone dosage due to the adsorption and catalytic ozonation.

3.4. Economic Analysis

The cost of this test mainly included ozone, electric power and catalyst. With 40 g/h ozone dosage and 0.2 m³/h influent flow, the ozone dosage of this system was calculated to be 40 (g/h) / 0.2 (m³/h) = 0.2 kg/m³. The ozone production price was about ¥ 11.1 /kg (\$ 1.58 /kg). Therefore, the ozone cost was ¥ 2.21 /m³ (\$ 0.316 /m³). The electric power consumption of various pumps, agitators, aerators and other devices was about 6.7 kW×h/m³. According to the electric price (¥ 0.602 /(kW×h)), the energy cost was about ¥ 4.06 /m³ (\$ 0.58 /m³). Including the additional cost (such as reagents, catalyst and water), the total cost of the system was about ¥ 6.65 /m³ (\$ 0.95 /m³), which was acceptable for the treatment of incineration leachate UF concentrate. During the operation of these devices, the actual power was lower than the rated power, so the actual operating cost was slightly lower than this cost above. However, this was still a rough analysis, and it would be better to consider the personnel costs, maintenance and initial investment, etc.

4. Conclusions

In this pilot-scale test, the ozonation-biological treatment-catalytic ozonation system was performed to remove total

nitrogen and organic matters from biological pretreated and membrane separated incineration leachate. Under the appropriate parameter conditions (influent flow of 0.2 m³/h, total gas flow of 0.5 m³/h, ozone dosage of 200 mg/L), the effluent COD was less than 50 mg/L. With the situation of high concentrated TN and low C/N, the experiment adjusted the influent ratio, simultaneously added composite carbon source and denitrifying bacteria, which ensure the effluent TN was lower than 20 mg/L. After repeated debugging, the combined system obtained 88.5% COD removal rate and 98.2% TN removal rate, which met leachate discharge standards in China (GB16889-2008). And the total cost was about ¥ 6.65 /m³ (\$ 0.95 /m³), which indicated that the system was practical in economics. Finally, the above-mentioned results indicated this combined system was more suitable for the treatment of refractory organics and highly-concentrated TN in leachate with a low COD_{bio}/N.

Acknowledgment

This work was financially supported by Shenzhen strategic emerging industry development special fund (JSCG20170823153043998).

Author Contributions

C.Y.M. (A graduate student) conducted all the experiments and wrote the manuscript. X.Y.T. (Professor) revised the manuscript. W.G.P. (Researcher), S.W. (Researcher), S.L.Q. (Researcher), C.Y.F. (Researcher), and M.A.C. (Researcher) conducted some experiments.

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