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Advanced nitrogen removal using bio-refractory organics as carbon source for biological treatment of landfill leachate

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ABSTRACT

A combined system was developed to enhance the chemical oxygen demand (COD) and total nitrogen (TN) removal from landfill leachate. The system consists of an up-flow anaerobic sludge blanket (UASB), anoxic/aerobic (A/O) reactor, and the denitrifying UASB (DUASB). The influent used in our study was a mixture of 1:2 raw leachate and domestic wastewater. Because of the inhibitory effect of free ammonia (FA) on the nitrite-oxidizing bacteria (NOB), stable partial nitrification occurred, with a consistent nitrite accumulation ratio in the A/O reactor. In the DUASB, the nitrite and nitrate nitrogen (NO_X^--N) were removed efficiently by denitrification by utilizing the carbon source of refractory organics under anoxic condition, which were likewise degraded. The organic matter, total nitrogen and ammonia nitrogen in the final effluent were 80–90, 39, and 14 mg/L, respectively, with respective removal efficiencies of 95, 98, and 95%. The respective COD removal efficiencies in the UASB, A/O and denitrifying-UASB were 76.8, 2.8, and 16.3%. The proposed process is an advanced technique that accomplishes the removal of organic matter and nitrogen in a single step.

1. Introduction

A large amount of leachate is generated when urban garbage is disposed of landfills. Currently in China, urban domestic garbage produces approximately 29 million tons of fresh leachate every year. Moreover, because of the high concentration of contaminants, a ton of leachate is equal to 100 tons of urban sewage. Landfill leachate is characterized by significant variations in water quality, complexity of composition, low carbon-to-nitrogen (C/N) ratio, poor biodegradability, high concentrations of organic matter and ammonia nitrogen (NH_4^+-N), and extremely expensive and difficult treatment. Consequently, the treatment of landfill leachate has become a topic of critical importance in scientific research [1–3].

Currently, the approach toward treating landfill leachate is mainly physical treatment, chemical oxidation [4–6], and bioprocessing [7–9]. Of which, bioprocessing is considered the mainstream method. Compared with the other approaches, bioprocessing by the nitrification-denitrification process or anaerobic ammoxidation can remove both the organic matter and the nitrogen contained in landfill leachate.

However, because of the peculiar characteristics of landfill leachate, the existing bioprocessing technologies, relevant to their economic viability and efficiency, are not able to remove the highly concentrated organic matter and nitrogen content. Mature landfill leachate, in particular, is characterized by a lower concentration of organic matter, increased humic substances, a higher concentration of NH_4^+-N, a low C/N ratio, a reduced 5-day biochemical oxygen-to-chemical oxygen demand (BOD_5/COD) ratio (from 0.4–0.6 to <0.2), and poor biodegradability. Therefore, the removal of refractory organics and the lack of a carbon source for the denitrification process are the major technological difficulties associated with bioprocessing. According to Peng et al. [10], if bioprocessing were used only, the effluent water COD would still contain some refractory organics at concentrations up to 500–1000 mg/L, which exceeds the prescribed discharge standard of China.

In order to increase the removal of refractory organics, certain post-processing methods are used in the stages subsequent to the bioprocessing stage, including a two-membrane process (ultrafiltration plus reverse osmosis), advanced oxidation, and coagulation. For example, Shrawan et al. [11] used the Fenton

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method to degrade the refractory organics contained in landfill leachate. However, this method is expensive, as there is a considerable demand for the Fenton reagent, which implies that this method is unsuitable for large-scale application.

In addition, landfill leachate has high ammonia nitrogen content; therefore, the total nitrogen content in the effluent water tends to be higher than the prescribed standard of China. In various studies, nitrification and denitrification have been employed as the main methods for removing nitrogen [12]. A number of studies have demonstrated that the nitrification of NH_4^+-N could achieve satisfactory results. However, there are various constraints to denitrification, including temperature, the biomass of the denitrifying bacteria, the availability of a carbon source, and the C/N ratio. Among these, a low C/N ratio and the lack of a carbon source are considered the primary constraints [13]. To improve the removal efficiency of total nitrogen and intensify the denitrification process, external carbon has to be added [14], which, obviously, implies additional expenses for operations, construction, and treatment. A recent study conducted by Zhang et al. [15] has demonstrated that refractory organics could be utilized as a source of carbon for denitrification; however, with two prerequisites, namely, an anoxic state and the availability of a large quantity of nitrates or nitrites. Accordingly, this study utilizes processes in which the nitrified liquid in oxic effluent water, which contains a large quantity of nitrates or nitrites, flows to an anaerobic reactor, where, subsequently, the refractory organics are thoroughly denitrified, providing a denitrification carbon source.

Additionally, to conserve the carbon sources, a partial nitrification process should be implemented by controlling the reaction conditions. Pochana and Keller [16] have found that COD at a concentration of 3.5–4.5 mg/(mg TN) is the carbon source required by the conventional biological denitrification. In contrast, for the partial biological denitrification process, the requisite carbon source was reduced by 40% and the aeration rate by 25% [17]. Consequently, the partial denitrification process is considered a good solution to the problem of the extremely low removal efficiency of total nitrogen in the treatment of landfill leachate.

Based on the understanding gained from the above-mentioned studies, we utilized a biological process, consisting of an UASB (referred to as UASB1), plus an anoxic/oxic (A/O) reactor and a denitrifying UASB (DUASB) system to treat leachate, generated by a municipal garbage landfill in Beijing, without using any physicochemical pre-treatment. A carbon source has to be provided to increase the removal of TN; however, in our process, the biorefractory organics were utilized as the carbon source for denitrification and there was no need to add external carbon. Consequently, in our process, the organic matter was used to the maximum extent. After part of the oxic effluent water, which consisted of nitrite and nitrate nitrogen (NO_X^--N), had flowed back into the UASB1, the organic compounds in the raw landfill leachate were depleted, as they had served as the carbon source for denitrification. After the oxic effluent water had flowed to the DUASB of the next section, the NO_X^--N were denitrified, and the refractory organics were depleted by serving as the carbon source. Therefore, this treatment process not only degraded the NO_X^--N contained in the oxic effluent water, but also thoroughly degraded the refractory organics contained in the system. Furthermore, partial nitrification was implemented in the A/O process, which removed the organics and total nitrogen economically and effectively. In addition, our experiment included factors that influenced the partial nitrification process.

2. Materials and methods

2.1. Test devices and operating conditions

As regards the test devices, a combined system, UASB-A/O-DUASB, was constructed for the experiment, as shown in Fig. 1. The UASB, DUASB, and A/O reactors and the clarifying tank were all made of organic glass, with the effective volumes being 8.3, 4.3, 15, and 20 L, respectively. The integrated feed tank was made of stainless steel. The A/O reactor was divided averagely into ten compartments, of which the first compartment was the anoxic zone, whereas the other compartments were oxic zones.

A mixture of raw leachate and domestic wastewater at a ratio of 1:2 was fed into the integrated feed tank. The influent used for the system was a liquid mixture that included the raw wastewater in the integrated feed tank and a small volume of effluent water from the clarifying tank (recycled nitrified liquid). After the liquid mixture had entered the UASB1, the NO_X^--N in the recycled nitrified liquid was denitrified by utilizing the abundant organic carbon sources contained in the raw leachate, while a small amount of

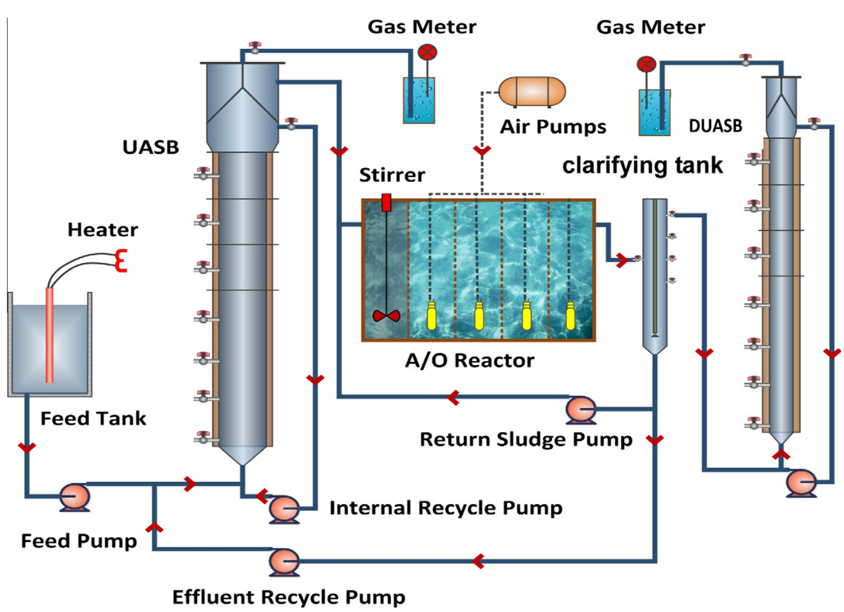

Fig. 1. Flow diagram of the UASB-A/O-DUASB system.