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Nitritation–denitritation in landfill leachate with glycerine as a carbon source

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HIGHLIGHTS

- Nitrogen removal from municipal landfill leachate via nitritation–denitritation.
- At limited oxygen concentration nitritation–denitritation occurred concurrently.
- Glycerine (Gly) may be successfully used as carbon source for denitritation.
- Gly increase in the total carbon source caused decrease in biomass production.

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ABSTRACT

The effects of limited oxygen concentration (0.7 mg O_2/L) in the aeration phase of the SBR cycle and glycerine as an additional carbon source on the effectiveness of nitritation–denitritation and sludge production during municipal landfill leachate treatment were examined. As carbon sources, sodium acetate (Ac) and sodium acetate (Ac) with glycerine (Gly) in the proportions of 3:1 (v/v) and 1:1 (v/v) were added. Low dissolved oxygen concentration inhibited the second stage of nitrification and nitrites were the main final products. Nitritation effectiveness was ca. 98–99%. Denitritation efficiency was relatively low (61%) in the reactor fed with Ac, which may be linked with high sludge production (Y_{obs} – 0.6 mg VSS/mg COD). Glycerine addition (Ac:Gly 1:1, v/v) caused an increase in process efficiency to 75.6% with a concurrent significant decrease in biomass production (Y_{obs} – 0.46 mg VSS/mg COD).

1. Introduction

Landfill leachates are considered wastewater that is difficult to treat since they undergo continuous qualitative and quantitative changes with landfill aging. Leachates from stabilized landfill contains high levels of ammonium, often reaching several thousands of milligrams per liter (Liang and Liu, 2008; Zhu et al., 2013) and a low concentration of biodegradable organics. Therefore, biological nitrogen removal via denitrification, proceeded by nitrification, is hindered by a low C/N ratio and to obtain high process effectiveness an external carbon source is needed. To date, the most popular are commercially available carbon sources such as methanol, ethanol or acetic acid, although this generates additional treatment costs. In order to eliminate this problem, two differentiated approaches may be applied: (i) technological solutions involving processes based on partial nitrification (oxidation of ammonia nitrogen to nitrite; nitritation) in combination with a short-cut denitrification (denitritation) (Fux et al., 2006; Aslan and Dahab, 2008) and (ii) the use of waste products as a potential carbon source for denitrification (Tora et al., 2011; Frison et al., 2013).

In the presented experiment, it was proposed that nitrogen removal from municipal landfill leachate would be due to both above-mentioned approaches, i.e. nitritation–denitritation and glycerine – waste-product from biodiesel production as a carbon source.

Partial nitrification requires a reduction in the activity of the nitrite oxidizing bacteria (NOB), without affecting the ammonia oxidizing microorganisms (AOB). One way to achieve this is to utilize the difference in the activation energies between ammonia oxidation (68 kJ/mol) and nitrite oxidation (44 kJ/mol). In the case of ammonia the higher activation energy means that the process rate can be made temperature-dependent (Schmidt et al., 2002). Further parameters conducive to short-cutting nitrification may be pH and dissolved oxygen (DO) regulation. However, in case of pH, discrepancies can be noted among the existing data (Villaverde et al., 1997; Ruiz et al., 2003; Wang et al., 2007). At low DO concentrations ammonia oxidizing bacteria are known to have a higher affinity for oxygen than nitrite oxidizing bacteria. Therefore the

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first phase of nitrification dominates (Schmidt et al., 2003; Zeng et al., 2009; Yang and Yang, 2011). So, in presented study, a control strategy to obtain nitritation was to adjust the air supply in the aeration phase to attain the required low dissolved oxygen concentration. Earlier studies concerning reject water treatment in SBR proved that this strategy is efficient for obtaining nitrite as a final nitrification product (Bernat et al., 2012, 2013).

The possibility of nitrogen removal from wastewater with a low C/N ratio is currently a subject of interest to many researchers. However, most of the works focus only on the process efficiency (Xu et al., 2012). It is known that in wastewater treatment by activated sludge, new cells (sludge) are one of the final products. Currently, with increased restrictions in sludge reuse and disposal, sludge treatment has become more challenging and more costly. Thus, an ideal approach to the sludge problem would be to reduce the excess sludge in wastewater treatment rather than post-treat the produced sludge. To reduce the production of biomass, the wastewater process must be engineered to divert the substrate from assimilation for biosynthesis to fuel exothermic, non-growth activities. For example, Abbassi et al. (1999) showed that a reduction of the excess sludge production by about 25% can be achieved by raising the oxygen concentration from 2 to 6 mg O_2/L in the mixed liquor. Partial nitrification is, on the other hand, observed at low DO concentration. Hence, an assessment of biomass production at low oxygen concentrations is needed. Similarly, it is important to check the effect of the use of waste-products as the external carbon source on biomass production. The few available studies concerning Y_{obs} value were carried out with commercial, pure carbon sources (Majone et al., 2001).

The main goal of this research was to determine the effectiveness of nitritation–denitritation during landfill leachate treatment in the SBR at a low DO concentration in the aeration phase using glycerine as the external carbon source. Moreover, sludge production under applied operational conditions was assessed.

2. Methods

2.1. Landfill leachate

Leachate used in the experiment originated from a well-organized landfill in Wysieka, near Bartoszyce (Warmia and Mazury district). The landfill site has been operating since 1996. Physico-chemical composition of the leachate was as follows: pH 8.12 ± 0.22; COD 732 ± 36 mg O_2/L; BOD_5 51 ± 6 mg O_2/L; BOD_5/COD 0.07; BOD_5/TKN 0.09; TKN 420 ± 28 mg TKN/L; 340 ± 14 mg N–NH_4/L; 48.9 ± 6.8 mg P/L; total dissolved solids 7032 ± 592 mg/L; volatile dissolved solids 1087 ± 126 mg/L, Cr 0.081 ± 0.025 mg/L; Cd 0.132 ± 0.26 mg/L; Cu 0.07 ± 0.002 mg/L; Ni 0.03 ± 0.0013 mg/L; concentrations of Pb and Hg were below detection limit.

2.2. Process configuration and system design

Experiments were carried out in three SBRs (SBR 1, SBR 2, SBR 3) with a working volume of 5 L each. Reactors were equipped with a stirrer with a variable speed control system (stirrer speed was maintained at the level 36 rpm). Dissolved oxygen was supplied using porous diffusers (fine bubble aeration was used), placed at the bottom of the reactors.

Moreover, the reactors were equipped with an oxygen concentration control system assuring a DO concentration in the aeration period of 0.7 ± 0.2 mg O_2/L. The reactors operated in a 24 h-cycle mode. Each cycle consisted of the following phases: filling (5 min), mixing (3 h), aeration (20 h), settling (50 min) and decantation (5 min).

The volumetric exchange rate was 0.3 d^{-1}. The system was operated at room temperature (20–22 °C) for 3 months.

Reactors were fed by the landfill leachate with the addition of an external carbon source in form of sodium acetate (Ac) and glycerine (Gly) (COD_{ext}). The dosage of the external carbon source should provide the COD_{ext}/TKN at the beginning of the reactor cycle ca. 4.0.

The chemical composition of glycerine was as follows: glycerol 80–85%, ash (NaCl) <7%, M.O.N.G. (matter organic non glycerol) <2%, methanol <0.5%, water – balance (product specification from Biodiesel Manufacturing Plant, Poland). Two solutions, one of sodium acetate and a second of glycerine were prepared in the following way: 150 g CH_3COONa or 113 g of glycerine was dissolved in 1 L of distilled water which resulting in 100 mg COD/ml each. The mixture of Ac and Gly, as a feed for SBR 2 and SBR 3, were prepared in the volumetric proportion. SBR 1 was fed only by Ac, whereas SBR 2 and SBR 3 by Ac and Gly in volumetric proportion 1:3 (v/v) and 1:1 (v/v), respectively.

2.3. Chemical analyses

Daily measurements of pollutant concentration in the effluent from the reactors included: chemical oxygen demand (COD), total Kjeldahl nitrogen (TKN), ammonia nitrogen, nitrites and nitrates. The activated sludge was analyzed for total suspended solids (TSS) and volatile suspended solids (VSS). In steady-state conditions, the measurements of COD, TKN, ammonia nitrogen, nitrites and nitrates during the SBR cycle were done. The analyses were performed according to APHA (1992).

3. Results and discussion

3.1. Organics and nitrogen removal kinetic

In the landfill, the leachate organic compound concentration (expressed as COD) was 732 mg COD/L. However, a low BOD_5/COD ratio (0.07) indicated that organics were persistent for biological treatment. The leachate originated from a landfill that has been in use for over 15 years and, therefore, showed a good correlation with the low organic compound content as low concentrations of organics in leachate from stabilized landfills is well-documented in the literature. However, recent investigations have revealed that even the leachate from young landfills contains low concentrations of organics. That may be caused by leachate recirculation which reduces the waste stabilization time, enhances gas production, and, consequently, lowers the leachate concentration, especially in terms of COD (Chan et al., 2002).

Apart from refractory organics, landfill leachate contains a high nitrogen concentration. Landfilled municipal solid waste (MSW) contains high amounts of organic nitrogen in a non-degradable form as well as readily-soluble nitrogen. As a result of anaerobic digestion of MSW putrescibles, around 50% of the nitrogen undergoes solubilization (Jokela and Rintala, 2003). Due to hydrolysis of soluble protein amino acids, dipeptides or oligopeptides are formed. Fermentation of amino acid leads to the formation of organic acids and ammonia. Leachate from older landfills is rich in ammonia nitrogen due to hydrolysis and fermentation of the nitrogenous fractions of biodegradable substrates. Ammonia concentration in leachate from different landfills may vary from tens or hundreds of mg N–NH_4/L (Statom et al., 2004) to 2000–3000 mg N–NH_4/L (Timur and Özturk, 1997).

In the presented study, the leachate contained mainly refractory organics and a high concentration of TKN (420 ± 28 mg/L). Therefore, in order to improve nitrogen removal, the external carbon