



Biological performance and sludge filterability of anaerobic membrane bioreactors under nitrogen limited and supplied conditions

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ABSTRACT

The impact of nitrogen on biological performance and sludge filterability of anaerobic membrane bioreactors was investigated in two lab-scale cross-flow anaerobic membrane bioreactors that were fed with cheese whey at two different COD:TKN ratios (50 and 190). Nitrogen deprivation adversely affected the biological treatment performance and reactor stability, as indicated by volatile fatty acids accumulation. On the other hand, nitrogen (urea) supplementation resulted in a reduced sludge median particle size and decreased sludge filterability. Standard filterability parameters such as capillary suction time and specific resistance to filtration tended to rapidly increase in the nitrogen supplemented reactor. The critical fluxes in the nitrogen limited and supplemented reactors were 20 and 9 L m⁻² h⁻¹, respectively. The rapid deterioration of sludge filterability under nitrogen supplemented conditions was attributed to abundant growth of dispersed biomass. Thus, the COD:TKN ratio of wastewater affected both bioconversion and filterability performance in the anaerobic membrane bioreactors.

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1. Introduction

Anaerobic membrane bioreactors (AnMBRs) are increasingly being researched in the last decade for the treatment of several wastewater streams due to their many advantages over conventional high rate anaerobic reactors (Dereli et al., 2012). They provide complete sludge retention, very high treatment efficiency, and excellent effluent quality free of suspended solids.

The achievable membrane flux in AnMBRs is governed by the cake layer covering the membrane, commonly referred to as membrane fouling, which results from the filtration process (Jeison and van Lier, 2007a; Charfi et al., 2012). In addition to the formation of a dense cake layer on the membrane surface, due to the accumulation of organic and inorganic foulants, clogging of membrane pores may contribute to membrane fouling in AnMBRs, resulting in a subsequent flux decline. Fouling is a very complex phenomenon, and its extent of manifestation depends on many parameters, such

as substrate characteristics, mixed liquor properties, bioreactor design and operating conditions, membrane properties and operation (Meng et al., 2009). Most of the time, it is difficult to identify a single parameter that determines the degree of fouling, because all these parameters are interrelated to each other (van den Broeck et al., 2011).

Among the causes and mechanisms of fouling, the impact of substrate composition is probably the least investigated one. In general, substrate characteristics have an indirect impact on fouling by affecting the bioreactor operating conditions, i.e. applicable organic load, food to mass ratio and hydraulic retention time, microbial species composition, characteristics of extracellular polymeric substances (EPS) and types of inorganic precipitates (LeClech et al., 2006). Organic matter and nitrogen content, which are generally referred to as carbon to nitrogen (C:N) or chemical oxygen demand to nitrogen (COD:N) ratio, of wastewaters is considered an important parameter affecting the performance of both aerobic and anaerobic biological treatment systems (Speece, 1996; Rittmann and McCarty, 2001). The COD:N ratio in aerobic membrane bioreactors (MBRs) is commonly investigated with regard to nitrogen removal and denitrification performance (Fan et al., 2014; Babatsouli et al., 2015). Obviously, a high COD:N ratio

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is in that case desirable, since an abundance of readily biodegradable substrate is needed in conventional denitrification systems. There are some contradictory reports about the effect of COD:N ratio on the membrane fouling and sludge filterability characteristics in aerobic MBRs. Sari Erkan et al. (2016) observed a decrease in sludge filterability indicated by lower critical fluxes in an MBR when the COD:N ratio of wastewater was increased. Controversially, several researchers indicated that low COD:N ratios promote fouling rates, deterioration of filterability and higher biomass yields in MBR systems (Feng et al., 2012; Hao and Liao, 2015; Hao et al., 2016). Considering the previous research, it is clear that the COD:N ratio has a substantial impact on mixed liquor characteristics (Ye et al., 2011) which are in close relation to filterability and fouling in aerobic MBRs.

In addition to a biodegradable carbon source, the anaerobic digestion process requires a balanced nutrient cocktail in terms of macro- and micro-nutrients that are required for bacterial and archaeal metabolism. Although the adverse effects of deficiency in micronutrients such as iron, nickel and cobalt have been well documented in literature (Speece, 1996; Demirel and Scherer, 2011; Hendriks et al., 2017), the impact of macronutrient limitation, such as nitrogen (N) and phosphorus (P) in anaerobic treatment is less well documented and macronutrient dosing is generally linked to the COD concentration and composition (van Lier et al., 2008). In general, the nitrogen and phosphorus demand for cell synthesis is low in anaerobic systems, due to very little biomass yield of anaerobic sludge. Speece (1996) reported that substrate COD:N ratio should be 50 and 150 for highly and lightly loaded systems, respectively. Others link this ratio to substrate composition and the expected growth yield, giving a COD:N:P ratio of 1000:5:1 for fully acidified wastewater and 350:5:1 for non-acidified wastewater (Chernicharo, 2007).

Several agro-industrial wastewaters, such as pulp and paper, olive mill, biodiesel production, confectionary and opium alkaloids industry effluents, are known to be nitrogen limited (Astals et al., 2011; Ersahin et al., 2011; Ozgun et al., 2012). In most of these cases, anaerobic processes are considered the most suitable technology available for the treatment of these high strength industrial streams. Owing to a reduced hydraulic selection pressure, anaerobic high-rate reactors are often limited by biomass wash-out when treating concentrated wastewaters. Particularly for these type of wastewaters, AnMBRs are of potential interest (Dereli et al., 2012), because the membrane can act as an absolute barrier against biomass wash-out and produce solids free effluent with high reuse potential. However, there is very limited information about the effect of nitrogen limitation (high COD:N ratios) on anaerobic treatment systems in particular AnMBRs. Nitrogen limitation in AnMBRs may exert a dual effect impacting the overall performance of the system. On the one hand, nitrogen limitation will restrict sludge production and may thus positively impact the membrane filtration performance. On the other hand, it may negatively affect the overall biochemical conversion process, leading to lower COD removal efficiencies. Nitrogen deficiency will likely induce metabolic changes in microorganisms and limit biomass synthesis. Consequently, sludge production and sludge characteristics will change and product formation and species distribution will be altered. Both will have consequences on treatment efficiency and reactor stability but also on sludge filterability and fouling propensity in AnMBRs.

Sam-Soon et al. (1990) reported that nitrogen deficiency caused poor formation of methanogenic sludge granules in UASB reactors. Controversially, Punal et al. (2000) observed an enhanced biomass adhesion in the start-up phase of anaerobic filters fed with nitrogen limited substrate. The adverse effect of nitrogen deficiency on the biological performance of AnMBRs was first mentioned by Qiao

et al. (2013) for the treatment of coffee grounds waste with a C:N ratio of 23.7. In this case, although the feed contained high amounts of organic nitrogen, the retardation of protein degradation by tannins limited the ammonification of organic nitrogen. On the other hand, given the high-enough ammonium nitrogen concentrations, even without nitrogen supplement, in the reactor ($250\text{--}500\text{ mg L}^{-1}$) for biomass growth, nitrogen deficiency seems unlikely. Therefore, the most plausible reason for poor reactor stability seems to be micronutrient deficiency as stated by the authors.

The purpose of this study is to investigate the effect of nitrogen limitation on both biological performance and sludge filterability in AnMBRs. According to authors' knowledge this is the first study systematically reporting about its effects in AnMBRs. Two AnMBR systems were operated with nitrogen limited and supplemented cheese whey. The sludge filterability was systematically evaluated under two different COD to total Kjeldahl nitrogen (COD:TKN) ratios with standard parameters in order to achieve an objective comparison.

2. Materials and methods

2.1. Reactor setup and operation

Two lab-scale cross flow AnMBRs with 10 L effective volume were operated under mesophilic conditions. Reactors were equipped with tubular ultrafiltration membranes (Pentair X-Flow) with a pore size of $0.03\ \mu\text{m}$. Membrane surface area was $0.014\ \text{m}^2$. A cross-flow velocity of $0.5\ \text{m s}^{-1}$ was imposed with a peristaltic pump (Watson Marlow 530U) and the permeate suction and backwash was conducted with a small sized peristaltic pump (Watson Marlow, 120U). A detailed schematic diagram of the reactors was previously presented (Dereli et al., 2014a). Daily biogas production and pH data were recorded online. The pH of the reactors was controlled with a stand-alone controller (Hach Lange SC-1000) and a dosing pump (KNF Stepdos O8 RC) for caustic addition.

The reactors were named R-1 and R-2 and were operated for 158 and 169 days, respectively. R-1 was fed with nitrogen limited substrate for 134 days and at the last stage nitrogen was added to the feed. R-2 operation was started with nitrogen supplemented substrate and nitrogen addition was first sharply and then gradually decreased within 40 days at the final operation phase. Switching of substrate towards the end of operation was applied as a control experiment in order to test and validate the effect of nitrogen limited and supplied conditions on the bioreactor performance. Both reactors had more than sufficient total phosphorus for biomass growth and COD:TP ratio was in the range of 70–75.

2.2. Experimental methods

2.2.1. Analytical methods

Chemical oxygen demand (COD) and total phosphorus (TP) concentrations were determined with Hach-Lange Kits. Total suspended solids (TSS), volatile suspended solids (VSS), total kjeldahl nitrogen (TKN) and ammonium nitrogen ($\text{NH}_4^+\text{-N}$) were measured according to Standard Methods (APHA, 1998). Soluble parameters were measured after centrifuging the sludge at $17,500\ \text{g}$ for 10 min and subsequently filtering the supernatant with $0.45\ \mu\text{m}$ disposable filters. Volatile fatty acids (VFAs) were determined with a gas chromatograph according to Dereli et al. (2015a). Extracellular polymeric compounds (EPS) were extracted and measured according to the methods described in Dereli et al. (2015b). Each sample was measured in duplicate.

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