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Nitrogen recovery from pig slurry in a two-chambered bioelectrochemical system

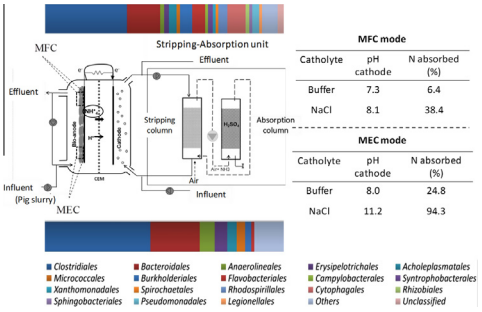
A. Sotres, M. Cerrillo, M. Viñas, A. Bonmatí *

IRTA, GIRO Joint Research Unit IRTA-UPC, Torre Marimon, ctra. C-59, km 12,1, E-08140 Caldes de Montbui, Barcelona, Spain

HIGHLIGHTS

- Ammonia migration is enhanced under higher organic and nitrogen loading rates.
- MEC mode, under higher voltages and NaCl presence, favored ammonia migration.
- NaCl promotes the stripping/absorption process in the cathode.
- Ammonia recovery was feasible using pig slurry as feed for the stripping-BES system.
- Shifting to MEC mode promotes changes in the Eubacterial and Archaeal community.

GRAPHICAL ABSTRACT



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ABSTRACT

Abiotic batch experiments showed that ammonia migration from anode to cathode was favored by an increase in voltage, from 39.9% to 44.6%, using synthetic media. A slight increase in ammonia migration was observed when using pig slurry, reaching a maximum of 49.9%. In a continuously MFC fed with pig slurry with a stripping/absorption unit coupled to the cathode chamber, the highest nitrogen flux (7.2 g N d^{-1} m^{-2}) was achieved using buffer as catholyte. Nitrogen flux increased to 10.3 g N d^{-1} m^{-2} when shifting to MEC mode. A clear improvement in nitrogen flux (25.5 g N d^{-1} m^{-2}) was observed when using NaCl as catholyte. Besides, ammonia stripping was favored, reaching a nitrogen recovery of 94.3% in the absorption column, due to the high pH reached in the cathode. The microbial community analysis revealed an enrichment of certain taxonomic Eubacterial and Archaeal groups when the system shifted from MFC to MEC mode.

1. Introduction

A conventional electrochemical system consists of a chemical anode and a chemical cathode separated by an ion exchange membrane, where the oxidation and reduction reactions take place in each compartment respectively. Bio-electrochemical systems (BESs) came upon the discovery of electrochemically active microorganisms which can catalyze the oxidation/reduction reactions being able to transfer the electrons to a solid surface such as the electrode (Pant et al., 2011; Clauwaert et al., 2008; Rabaey et al., 2010). A BES is considered a microbial fuel cell (MFC) if it gathers electrical power, and is considered microbial electrolysis cell (MEC) if electrical energy is supplied to drive an otherwise non-spontaneous reaction (Hamelers et al., 2010).

One advantage of a BES with a two chamber configuration is that the oxidation and reduction products are produced in separated compartments, making possible to recover "clean" products

* Corresponding author. Tel.: +34 902 789 449; fax: +34 938 650 954.
E-mail address: august.bonmati@irta.cat (A. Bonmatí).

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out of wastes (Hamelers et al., 2010). For this reason, BES are increasingly becoming a novel technological solution for environmental issues related to wastewater treatment, nutrient recovery, and the production of valuable compounds, with the advantage that they can help to reduce energy costs.

Nitrogen is one of the key contaminants and its discharge regulation is becoming stricter. In relation to nitrogen management, there is a dichotomy between strategies based on transferring nitrogen to the atmosphere (N_2) through biological nitrification–denitrification (NDN), and strategies that focus on the nitrogen recovery by means of producing struvite or concentrated nitrogen streams that are potentially reusable in agricultural areas with nutrient shortages: stripping-absorption (Bonmatí and Flotats, 2003a), thermal concentration (Bonmatí and Flotats, 2003b; Bonmatí et al., 2003), struvite precipitation (Cerrillo et al., 2015). However, to develop or further improve a technology for nitrogen removal/recovery is becoming a challenge in BES research. It is well known that nitrogen can affect the BES performance since the concentration of total ammonia nitrogen (TAN) can inhibit the activity of the microbial community, and accordingly can affect the anodophilic population reducing electricity generation (Nam et al., 2010). Nevertheless, and as it is reported that microorganisms can acclimatize to increasing TAN concentrations, the inhibition concentration threshold is not clear. In this sense, Kim et al. (2011) reported a higher TAN inhibition threshold in MFCs operating in continuous mode, than in batch mode. Besides ammonia inhibition depends on the anolyte pH (Kuntke et al., 2011). With a low anolyte pH, an increase in ammonia concentration does not affect the performance of a MFC in the same way, owing to the low anolyte pH which results in less free ammonia. Therefore, understanding how nitrogen can affect a BES performance is a key factor to improve strategies for nitrogen removal/recovery from organic wastes.

So far, different processes have been studied for nitrogen removal/recovery using BES technologies. It has been reported that nitrogen can be removed by using biological processes such as external nitrification and subsequent denitrification, accomplished by microorganisms in the cathode chamber (Clauwaert et al., 2007; Virdis et al., 2008), or even with simultaneous nitrification–denitri fication in the cathode (Virdis et al., 2010). Another strategy for nitrogen recovery is to combine biological and physicochemical processes. The fundament of this strategy in a BES system is the fact that ammonium ions can diffuse from the anode onto the cathode compartment throughout the cation exchange membrane (Rozendal et al., 2008), either via diffusion or current-driven migration (Kuntke et al., 2011). Then, the ammonia can be recovered by means of stripping, promoted by active aeration and a high catholyte pH, and its subsequent adsorption in an acid solution. This strategy has been previously described by Kuntke et al. (2012), who developed a MFC reactor to simultaneously produce energy and recover ammonium from real urine, reaching an ammonium recovery rate of 3.29 g N d^{-1} m^{-2}, at a current density of 0.50 A m^{-2}. Later on, Desloover et al. (2012) developed a non biological electrochemical cell in which a stripping unit was coupled to the cathode chamber, reaching a nitrogen flux of 120 g N d^{-1} m^{-2}, with synthetic wastewater. Although this technology has already been reported as a sustainable process to recover ammonia from wastewater, further research is needed when applied this strategy to treat high strength animal wastewater, such as pig slurries, aiming to accomplish the requirements needed for agricultural uses, and equally to investigate more in depth its performance in MFC and MEC modes.

Setting up a biofilm on the electrode surface is essential for efficient performance in a BES (Franks et al., 2010). Although both MFCs and MECs favor the growth of exoelectrogenic bacteria, microbial communities in MECs may be different from those in MFCs, because distinct conditions favor the growth of different microorganisms, as previously described (Logan et al., 2008). So far, there is a lack of knowledge about the best method for enriching an exoelectrogenic microbial community, but a common practice described by Ditzig et al. (2007) is to switch from MFC mode to MEC mode for the inoculum to be initially acclimated and for an exoelectrogenic community to be preselected in the anode of the MFC.

In this work, the use of both synthetic wastewater and the liquid fraction of pig slurry and the effect of the applied voltage on ammonia recovering, were studied. Furthermore, the feasibility of nitrogen recovery using a stripping unit coupled to the cathode of a continuously fed BES was investigated. Whereas the stripping system was operated under MFC and MEC modes, a third goal was to analyze the effect of shifting from MFC mode to MEC mode on the microbial community (Eubacteria and Archaea) of the bio-anode, by means of 454-pyrosequencing.

2. Methods

2.1. Experimental set-up

The experimental set up consisted of a two-chambered bioelectrochemical system (BES). The BES was built using methacrylate plates, with the anode and cathode compartment ($0.14 \times 0.12 \times 0.02$ m^3) separated by a cation exchange membrane CEM (14×12 cm) (Ultrex CMI-7000, Membrane International Inc., Ringwood, NJ, USA). The anode frame had a window which allowed taking samples from the biomass attached to the electrode. A carbon felt mesh, 3.18 mm thick, 99.0% Carbon (Cymit Química, S.L.) and stainless steel mesh, were used as anode and cathode electrodes respectively. Before use, organic and inorganic impurities were eliminated from the carbon felt sequentially soaking it in HCl 1 M and NaOH 1 M, each time for 24 h, and then rinsing it with deionised water. The projected anode surface area was of 0.0209 m^2. Copper wires were used to connect the electrodes to an external 500 Ω resistance (MFC mode) or to a power supply unit (MEC mode).

When the BES was fed with the liquid fraction of pig slurry a stripping/absorption unit was coupled to the cathode compartment, following to the set up previously described by Desloover et al. (2012). The system consisted of two tubular columns (height: 70 cm, external diameter: 7 cm, internal diameter: 5.5 cm) both filled with Raschig rings (length: 5–7 mm, internal diameter: 3 mm, external diameter: 5 mm). The stripping column was filled with 80 ml of medium from the cathode chamber and the absorption column with 500 ml of H_2SO_4 1 M. The spraying of the catholyte over the stripping column was completed installing a Mini diaphragm vacuum pump (VP 86, VWR International) at the bottom of the column; a water trap was also connected to protect the pump.

2.2. Batch experiments

Batch experiments were performed at room temperature (23 ± 1 °C) in abiotic conditions.

The anode feed consisted of either synthetic wastewater or the liquid fraction of pig slurry with an ammonium concentration of 523 mg N-NH_4^+ L^{-1}; and a buffer solution or sodium chloride for the cathode compartment, depending on the experimental design. All abiotic experiments were performed under different voltages: passive diffusion (P.D.), 0.3 V and 0.6 V, using a power supply unit (PS23032, Diotronic, S.A.). Each experiment lasted 56 h and every condition was tested in triplicate.