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Domestic wastewater treatment in parallel with methane production in a microbial electrolysis cell

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ABSTRACT

Microbial electrolysis cells (MECs) have great potential as a technology for wastewater treatment in parallel to energy production. In this study we explore the feasibility of using a low-cost, membraneless MEC for domestic wastewater treatment and methane production in both batch and continuous modes. Low-strength wastewater can be successfully treated by means of an MEC, obtaining significant amounts of methane. The results also suggest that hydrogenotrophic methanogenesis reduce the incidence of homoacetogenic activity, thus improving the overall MEC performance. However, gas production rates are low and important aspects such as methane solubility in water still remain a challenge. Overall, MECs can offer competitive advantages not only for low-strength wastewater treatment but also as an aid to anaerobic methane production by improving the chemical oxygen demand (COD) removal and methane production rates.

1. Introduction

Domestic wastewater (WW) usually requires treatment before being discharged into the environment. Activated sludge, a widespread WW treatment process, is an energy-intensive technology that demands ~1% of the total Spanish electrical energy consumption [1]. WW may contain significant amounts of chemical energy stored in the dissolved organics (some authors have estimated 17.8 kJ g^{-1}-COD [2]) that, if appropriately recovered, could improve the energy balance of the process. This residual energy content can be exploited by means of biological treatments, among which anaerobic digestion (AD) is preeminent. AD has proved to be a reliable, flexible and robust technology to treat and valorize a wide range of organic waste streams, but it usually fails to treat low-strength WW.

Microbial electrolysis cells (MECs) are a leading edge bio-based technology that has recently emerged as an alternative to conventional processes for organic waste treatment and energy production. In contrast to AD, MECs can treat diluted wastewater streams at relatively low temperatures [3], and have already delivered promising results in the laboratory [4] and in studies at semi-pilot [5] and pilot scales [6]. MECs are usually aimed at producing hydrogen gas (as opposed to the methane produced by AD), a fuel with a high gravimetric energy content and an important feedstock in many industrial processes. However the production of hydrogen presents numerous challenges (especially when treating low-strength WW) that jeopardize the upscaling process [7]. Indeed, if the hydrogen produced at the cathode is not readily evacuated it can be re-oxidized at the anode, a phenomenon known as hydrogen recycling, which deteriorates the performance of the MEC by limiting COD removal and reducing energy recovery [8]. In addition, cathodic hydrogen can be converted to acetate by homoacetogenic microorganisms resulting in a phenomenon with similar consequences to hydrogen recycling (i.e. limited COD removal and energy recovery [9,10]). These difficulties could easily be overcome by using polymer membranes to separate the anolyte from the catholyte. However, polymer membranes are usually expensive, make the reactor design more complex, and increase the energy usage of the MEC, which finally impacts the capital and operational costs and threatens the prospect of practical application of this technology.

Aside from interfering in the bioelectrochemical operation of the MEC, hydrogen management presents additional challenges. For instance, due to its low volumetric energy content, if hydrogen is not used *in situ* in the WW treatment facility, it would require intensive compression for its transportation, an operation that

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demands a substantial amount of energy [11]. Moreover, the equipment required for compression is usually expensive [11], and may not always be justified for the low hydrogen productivity of MECs [12]. Finally, the hydrogen produced in membraneless MECs is usually mixed with significant amounts of methane, which complicates its use as a feedstock for high-added-value industrial applications (that usually require high purity hydrogen), thus restricting its use for energy valorization. This contamination is hard to avoid since methanogenesis cannot be successfully suppressed once it becomes dominant in an MEC [13]. Moreover, it has recently been discovered that *Methanosaeta* and *Methanosarcina*, often the most abundant methanogens in anaerobic digesters, can accept electrons via direct interspecies electron transfer from *Geobacter metallireducens* [14–16], which increases the difficulty of finding a satisfactory solution for hydrogen contamination with methane.

Rather than avoiding its production, methane might prove to be a suitable alternative to hydrogen whenever the aim is to produce a reliable fuel from WW. If cathodic hydrogen can be converted to methane as soon as it is produced, or even if the electrons that arrive at the cathode could be used directly to reduce carbon dioxide to methane, the difficulties associated with homoacetogenic activity and the hydrogen recycling phenomenon would be ameliorated, if not completely removed, thanks to the presence of hydrogenothrophic methanogens [9]. This is because the Michaelis–Menten constant for hydrogenotrophic methanogens is significantly lower than for homoacetogens, and the H_2 threshold concentration for methanogens is also several orders of magnitude lower [17]. Moreover, methane can be produced in MECs with a relatively simple design and with the additional advantage that methane is an easier gas to manage than hydrogen.

In this study, we aimed to assess the opportunities for scalability of low-cost MECs for WW treatment and methane production, paying special attention to the problems associated with the use of low-strength WW where MECs offer a competitive advantage over aerobic WW treatments [18]. We also explore some of the benefits of using MECs to assist anaerobic methanogenesis.

2. Material and methods

2.1. Influent

Synthetic medium (SM) and real domestic wastewater (WW) were used as influents for the MEC at different stages of the experiment. The composition of both SM and WW are described in Table 1. WW was collected from the municipal wastewater treatment plant of León (Spain). The chemical composition of SM was (in mg L^{-1}): K_2HPO_4 (14.3); KH_2PO_4 (46.2); NH_4Cl (90); NaCl (20); $CaCl_2 \cdot 2H_2O$ (30); $MgSO_4 \cdot 7H_2O$ (100); yeast extract (10); and 1 mL L^{-1} of a trace metal solution of composition: $MgCl_2 \cdot 6H_2O$ (410); $MnCl_2 \cdot 4H_2O$ (50); $FeCl_2 \cdot 4H_2O$ (50); $NiCl_2 \cdot 4H_2O$ (12); $ZnSO_4 \cdot 7H_2O$ (10); $CoCl_2$ (7.7); $CaCl_2 \cdot 2H_2O$ (30); $Al(NO_3)_3 \cdot 9H_2O$ (29.4); Na_2SeO_4 (8.7); $Na_2MoO_4 \cdot 2H_2O$ (2); $CuSO_4 \cdot 5H_2O$ (1); H_3BO_3 (2); $NaWO_4 \cdot 2H_2O$ (1). The carbon source was sodium acetate (300 mg-COD L^{-1}). All solutions were filter sterilized and stored at 4 °C to prevent microbial growth. Distilled water was used for solution preparation, and the chemicals and reagents used were of analytical grade.

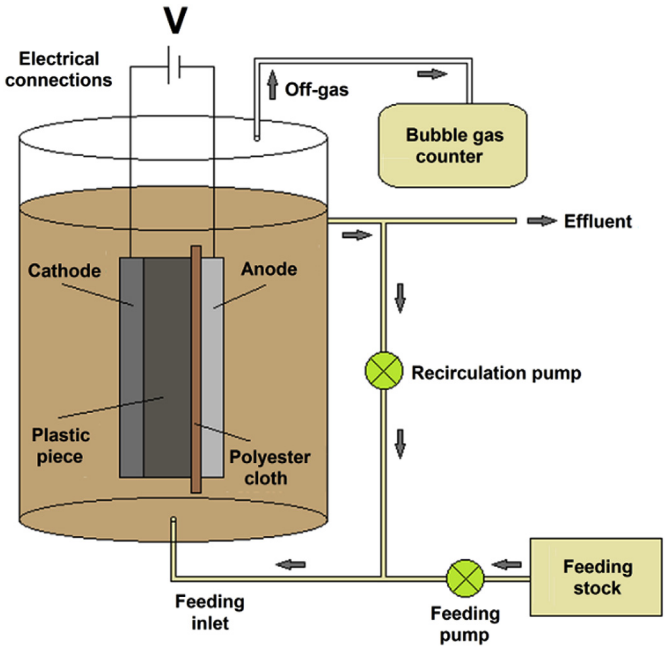

Fig. 1. Schematic representation of the laboratory set-up.

2.2. MEC design, instrumentation and operation

A continuous-flow, single-chamber, membraneless MEC, with a total volume of 3 L was used for the experiment (Fig. 1). The reactor was equipped with connections for gas exits and liquid entries and exits, and contained the electrolytic module (anode + cathode) submerged in the feeding stock, either SM or WW. The anode consisted of one layer of carbon felt 1 cm thick (Sigratherm soft felt GFD 2, SGL Carbon Group, Wiesbaden, Germany), and the cathode was a stainless steel electrode, both with dimensions of 210 × 100 mm. One 0.6-mm-thick piece of polyester cloth was sandwiched between the anode and the cathode to avoid any electrical contact between the two electrodes. The inter-electrode separation was set to 1 mm. Prior to its introduction in the reactor, the anode was inoculated with the effluent from another

Table 1
Characterization of the five sets of domestic wastewater used as influent in this study, and of the synthetic medium (ND = not determined).

	Domestic wastewater (WW)					Synthetic medium (SM)
	HRT 4 h	HRT 8 h	HRT 12 h	HRT 24 h	Batch	
TSS (mg L^{-1})	41.0 ± 1.0	45.8 ± 0.3	55.3 ± 1.8	53.8 ± 1.3	55.0 ± 0.5	ND
VSS (mg L^{-1})	9.8 ± 0.3	9.3 ± 0.3	10.0 ± 2.0	9.8 ± 0.8	10.8 ± 0.8	ND
TOC (mg L^{-1})	22.7 ± 1.4	24.6 ± 1.3	32.7 ± 1.6	34.0 ± 1.0	89.4 ± 11.9	168.8 ± 8.1
Total N (mg L^{-1})	19.7 ± 0.9	21.0 ± 1.0	20.5 ± 1.4	20.9 ± 1.2	59.4 ± 3.0	26.5 ± 1.8
NH_4 (ppm)	18.9 ± 5.1	20.9 ± 5.4	20.0 ± 6.2	19.1 ± 5.2	21.5 ± 4.9	36.5 ± 10.5
pH	7.08 ± 0.12	7.22 ± 0.12	7.01 ± 0.06	7.13 ± 0.10	7.16 ± 0.14	6.49 ± 0.12
Conductivity (μS cm^{-1})	504 ± 10	486 ± 12	498 ± 12	496 ± 12	492 ± 10	732 ± 14
COD (mg L^{-1})	65 ± 2	67 ± 3	77 ± 3	78 ± 3	188 ± 20	450 ± 22
Acetate (mg L^{-1})	36 ± 6	27 ± 2	33 ± 6	29 ± 5	27 ± 1	330 ± 24