IMPROVEMENT of BIOGAS GENERATION by COUPLING BIOELECTROCHEMICAL SYSTEMS to ANAEROBIC DIGESTION

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1. BACKGROUND and MOTIVATION of the PROJECT

The primary aim of the proposed research project is to define the possibility to control the performances of an anaerobic digester, in terms of methane production, through the installation of a bioelectrochemical system directly connected to the digester. Furthermore, while methane produced via currently established anaerobic digesters derives from high-strength wastewaters, generation of methane from diluted wastewaters can be achieved via bioelectrochemical systems: adding further potentials to the generation of biogas.

1.1 Background and Motivation

Bioelectrochemical systems (BES) are based on the ability of microorganisms to transfer electrons to solid state electrodes upon oxidation of organic substances. The most extensively studied BES is the microbial fuel cell (MFC). Microbial degradation of organic compounds can be seen in most cases as an oxidation, where the final acceptors of electrons from the nutrients is a soluble molecule, such as O₂, NO₃ or SO₄²⁻. In MFC, microorganisms switch from the natural electron acceptor to an insoluble acceptor, such as the anode. The electrons then flow through a resistor to a cathode, at which the electron acceptor, typically oxygen, is reduced (Fig. 1). Microorganisms in the anodic chamber oxidize organic substrates and generate electrons and protons through a dissimilative process. Electrons flow from the anode to the cathode through an external circuit, while protons cross a proton exchange membrane (PEM) and combine with oxygen to form water. The generation of electric current is made possible by keeping the microorganisms separated from electron acceptors other than the anode: as oxygen is a natural acceptor, an anaerobic anodic chamber is required [1].

![Figure 1. Schematic representation of a two-chamber MFC. Microorganisms in the anode chamber degrade organic substrates via oxidation and donate the liberated electrons to the anode. Electrons flow to the cathode through an external circuit, while protons (H⁺) pass through a selective proton exchange membrane (PEM).](image)

Currently, the most promising application of MFC is in connection to wastewater treatment process, where microorganisms present in wastewaters [2,3] can be exploited to both degrade organic compounds in wastewaters and generate electrical current at the same time. It is in fact thought that, upon optimization of the MFC system, the amount of electricity generated might at least be used to supply energy to the wastewater treatment plant and prevent the costs linked to aeration (as MFC requires an anodic anaerobic compartment), saving in this way 70% of the total costs typically borne by the plant [4]. In addition, the potentiality of a coupled anaerobic digester-MFC (AD-MFC) system has also been preliminarily shown, highlighting the possibility of removing the
sulphide generated in AD (98% sulphide was removed) via oxidation by controlling the anodic potential of the MFC. Therefore, such a system might be improved and implemented to clean AD effluents with concomitant electricity generation [5].

A modification of MFC defined microbial electrolysis cell (MEC) has been conceived to store electrical energy as biofuel (i.e. hydrogen, H₂). In a MEC, the cathode chamber is kept under anaerobic conditions, so that the electrons flowing to the cathode can combine with protons thus generating H₂. H₂ gas evolution at the cathode, however, is not spontaneous: the voltage produced by electrogenic microorganisms at the anode typically is not sufficient to evolve H₂ at the cathode. Therefore, a small external voltage (≥ 0.114 V) in addition to the one generated by microorganisms is necessary [6-8]. To summarize, MEC is an electrolysis reactor that produces H₂, while MFC is a fuel cell that produces electricity.

One of the main disadvantages of MEC is the requirement of expensive material as cathodic catalyst. Platinum would be the cathodic material of choice, thanks to its excellent electrocatalytic activity towards H₂ evolution. Therefore, the need of cheaper and more sustainable material has prompted research into alternative cathode catalysts. Such research led to the development of the so called microbial biocathodes, where microorganisms can grow and catalyse the transfer of electrons from the cathode to H⁺ (in the case of H₂ generation) or to other oxidized compounds. Even though the cellular and molecular mechanisms of the biologically catalysed electron transfer are still not completely clear [9], it has been shown in MFC systems that microbial biocathodes catalyse the electron transfer to electro positive terminal acceptors, such as O₂ or nitrate [10]. This biologically catalysed reduction to the generation of a biofuel (H₂) or other chemicals (e.g. H₂O₂) at the cathode [11] has been proposed to provide larger environmental benefits, compared to the generation of electric power via MFC [12].

Interestingly, it has also been demonstrated that electrons can be transferred from the cathode to CO₂, resulting into electrochemical reduction of CO₂ to methane (CH₄) according to the reaction:

\[ CO_2 + 8H^+ + 8e^- \rightarrow CH_4 + 2H_2O \] (1)

The electrochemical generation of methane is a rather recent application of MEC systems. One of the first demonstrations of the possibility to generate CH₄ exploiting the hydrogen produced at the cathode of a MEC was performed by Clauwert et al. [13]. In this study, the H₂ generated at the cathode via oxidation of acetate at the anode was bubbled into a lab-scale anaerobic digester. The study demonstrated that the cathodically produced H₂ could efficiently be converted into CH₄ at room temperature at a ratio of 0.41 mol methane · mole⁻¹ acetate.

While the CH₄ generation reported by Clauwert et al. [13] was shown by connecting in series a MEC and an AD, the possibility to produce CH₄ from CO₂ using an electrode as electron donor in a two chamber MEC has been shown by Cheng et al. [6]. The authors of this study have reported an electron capture efficiency of 96% at -1.0 V (applied voltage), which is much higher than the 10-57% efficiency achievable using metal catalysted electrodes for methane production [6]. Very similar results were obtained by Villano et al. [14], showing that the reduction of CO₂ was achieved most probably thanks to the presence of hydrogenophilic methanogens bacteria and that the rate of extracellular electron transfer was strongly dependent on the cathode potential.

Furthermore, the increase of the specific cathode surface is thought to additionally increase the rate of the overall electron transfer from the electrode to the microorganisms.
The interest in the development of MFC and MEC systems is not based on the possibility to use such systems for electricity production, as the values of electric power achievable cannot compete with already existing technologies. Nevertheless, BES are currently regarded as very promising technologies to be coupled to anaerobic digestion for biogas generation [6,13,15]; to different steps of waste water treatment [16-18] and to the treatment of polluted industrial streams and soils [19,20]. Therefore, deeper knowledge of the systems would lead to the optimization of BES, by defining the optimal structural and biological properties according to their specific applications. Overall, development of BES technologies means improvement of one further "green" technology to support and improve the outcomes of the currently established ones.

1.2 State of the Art

Microbial electrosynthesis (MES) via MEC systems is a rather novel research field and not yet largely developed on industrial scale. However, the growing production of electrical power from solar or wind energy has led to the renewed interest in energy storage or conversion technologies to solve transport and storage issues associated with electrical energy. MES can contribute to address these issues by allowing on site conversion of electrical energy (current) to chemical energy (fuel) [21]. The technology as such has large potential, as several examples for further electrochemical synthesis processes have been shown, including CO₂ reduction to multi-carbon compounds [22]; reduction of low value chemicals to added value chemicals (e.g. glycerol to 1,3-propanediol); generation of hydrogen peroxide (H₂O₂) and sodium hydroxide (NaOH) from waste streams [11]. The production of H₂O₂ and NaOH delivers substantial value return due to the high production rate and market price. Pilot-scale plants of such kind already exist and they actually demonstrate that it would be possible to move wastewater treatment into the bioproduction space.

To the best of our knowledge, there are no installations of AD-MEC systems on sites of biogas production, but the ongoing research has demonstrated on the lab-scale that production of methane using MEC systems is efficient and has a number of advantages, such as:

i) no need of thermal energy
ii) diluted waste streams can be efficiently used as substrates
iii) the system can be easily controlled via application of different voltages

However, very few studies have been performed on this kind of systems in relation to methane production and the best setup of an AD-MEC coupled system has to be defined. Therefore, the presented project will add precious and fundamental knowledge that will speed-up a future in situ application. The described advantages of electromethanogenesis will not anyway displace the already existing anaerobic digesters for biomethane production, but once the best AD-MEC setup would be determined, AD-MEC systems would guarantee higher methane productions, coupled to CO₂ capture and further waste stream treatment.

1.3 Previously featured research work in this field of research from the department or University

The research field on MFC at Chalmers University of Technology has been started and established by the applicant in 2010 thanks to a funding received from Göteborg Energi Forskningsstiftelsen (project nr. 09-13).
The project focused on the use of **metal alloys with unique foam structure** to be used as electrodes in MFC systems. The research performed determined that the foam structure of metal alloys represents an actual advantage for the performance of MFC compared to the use of massive electrodes, as a **fourfold increase of the current density at the anode has been achieved in MFC with stainless steel foam electrodes**. The project performed has represented an exploratory study, which has been crucial to define optimal conditions and ways to overcome problems that might be encountered while establishing MFC systems. Therefore, the experience gained will be of great advantage while establishing MFC and MEC systems with novel applications.

Furthermore, thanks to mediation of Göteborg Energi during the development of the project, the applicant has established direct connections with Gryaab AB (Göteborg, Sweden): the municipal wastewater treatment plant in Göteborg, which will still be a potential collaborator throughout the project proposed here.

Therefore, since metal foam electrodes allowed improving the MFC output, we think it is worth implementing this kind of electrodes also in AD-MEC systems, as the peculiar structure and materials could further ameliorate MEC performance as well, hence increasing the methane development at the cathode.

The project will be carried on in collaboration with the **Department of Civil and Environmental Engineering** at Chalmers University of Technology, where some preliminary studies have been performed regarding the potentialities of coupling an anaerobic digester to a MEC.

The experimental setup used in the preliminary experiments consisted of a 1.1-litre anaerobic digester connected to a 0.5-litre BES. The anaerobic sludge liquor was recycled through the anode and cathode chambers of the BES. The anode and cathode were both made of solid graphite. A voltage of 0.6 V was applied to the BES. Both the anode and cathode electrodes showed improved bioelectrochemical activity during the course of incubation. **Fig.2** below shows cyclic voltammograms of both electrodes on day 8 and day 17 of the experiments. We can see that the current density peaks are larger on day 17, suggesting both anode and cathode electrodes improved their performance over time. The idea is that a BES can be used to improve and control the performance of AD. The anode can be used to control the level of volatile fatty acids in the AD tank whereas the cathode can be used to stimulate biogas production. **Fig.2** shows that both anode and cathode exhibit the desired electrochemical activity. Further experiments are necessary to investigate the product conversions that occur at the respective electrodes and how these may affect AD performance.

![Figure 2. Cyclic voltammograms (5mV/s) of anode and cathode electrodes in a BES connected to AD. Cyclic voltammetry studies qualitative information about electrochemical processes under various conditions, such as the presence of intermediates in oxidation-reduction reactions, the reversibility of a reaction.](image-url)
The outcomes of project 09-13 have been presented in a few national and international occasions listed below:

“From waste to electricity – is microbial fuel cells the answer?”
Mapelli V.
Chalmers Energy Conference
27th January 2011, Chalmers University of Technology, Göteborg, Sweden.

“Metal foam electrodes and lignocellulosic waste in MFC set-up”
Mapelli V., Mapelli C., Iranmanesh P. and Olsson L.
3rd International Microbial Fuel Cell Conference 2011

“Viability study of the use of cast iron open cell foam as microbial fuel cell electrodes”
Mombelli D., Mapelli C., Mapelli V., Gruttadauria A., Baldizzone C.
MetFoam 2011
18th-22nd September 2011, Busan, South Korea

Use of metal foams as microbial fuel cell electrodes
Mapelli V., Mombelli D., Gruttadauria A., Baldizzone C. and Mapelli C.,
Manuscript in preparation

2. INDUSTRIAL RELEVANCE

2.1 Driving Forces
The main and general driving force of the proposed project is the possibility to decrease our dependence on fossil fuels via the establishment sustainable technologies for energy production. Bioelectrochemical systems (BES) are one of the attractive technologies able to generate energy in the form of electricity from waste and renewable sources. Even though such systems are not competitive with existing clean technologies for electricity generation, the added value of BES is their potential to couple generation of electric current to sustainable production of chemicals and biofuels and to decontamination of soils and waters.

The main specific driving force is the possibility given by microbial electrolysis cells (MECs) to generate methane via capture and reduction of CO₂ and to optimize the yields of methane production obtained via the established anaerobic digestion. MECs are not intended to substitute the existing mature technology for CH₄ production, but they can be directly integrated into existing infrastructures in order to improve the overall performance of the biogas production process.

In a system as the one designed in this project, factors that would contribute to increase the overall efficiency of biogas production are:

i) the further degradation of residual organics that is not sustained by conventional anaerobic digesters
ii) the possibility to produce methane in the MEC even at low substrate concentrations compared to the high-strength waste waters typically used in anaerobic digesters
iii) the possibility to remove residual sulphide from effluents from anaerobic digesters
iv) the possibility to control the system by tuning the external potential applied to the MEC.
2.2 Future Areas of Application

The proposed research aims at determining the possibility to improve the process of biogas ($\text{CH}_4$) generation in an anaerobic digester, by coupling it to a MEC. Preliminary results point towards this direction; however, a number of technical and biological factors have to be further studied in order to determine the optimal configurations. The outcomes of the project achievable on lab-scale will outline the critical features of the system to be taken into consideration when implementing AD-MEC on pilot- and real-scale.

A successful integrated system constructed on the basis of the one proposed in this project would both increase the methane production and capture the $\text{CO}_2$ with higher efficiency compared to the $\text{CO}_2$ capturing methods currently used based on metal catalysed electrodes. Furthermore, exploiting the potential of MEC systems, biogas in future could also be produced from sources that currently are not considered for methane production due to their low organics concentration, therefore, the MEC-produced biogas will contribute to increase the total amount of methane available as fuel from renewable sources.

3. AIM of the PROJECT and PROJECT DESCRIPTION

3.1 Actual Aims of the Project

- **Coupling of Microbial Electrolysis Cell to Anaerobic Digestion to improve methane production**
  Improve and control the production of biogas ($\text{CH}_4$) occurring in a conventional anaerobic digester (AD) by coupling it to a microbial electrolysis cell (MEC).

In a MEC, microorganisms thriving in the cathode chamber (biocathode) are able to transfer the electrons generated from microorganisms in the anode chamber and the ones supplied via external voltage application to $\text{CO}_2$, thus generating $\text{CH}_4$. The project aims at defining the critical factors that will make the implementation of AD-MEC systems as effective as possible.

- **Definition of critical factors and parameters towards optimization of AD-MEC performance**
  Characterization of an AD-MEC coupled system by testing different substrates, setups, electrodes and applied potentials. The knowledge gained through a basic characterization of the system will be crucial for actual implementations of the system on the field where AD are already settled and working.

3.2 Other (indirect) Aims of the Project

- **Identification of metal alloys with beneficial effects on MEC performance**
  The use of metal foam electrodes differing in composition is also a novel aspect of the research and will lead to the possible selection of metals that, being cheaper than platinum, might still have good properties as metal catalysts for electrical assisted $\text{H}_2$ generation and also allow the formation of the microbial biofilm on both electrodes.

- **Efficiency of $\text{CO}_2$ capture coupled to $\text{CH}_4$ generation**
  One of the indirect outcomes of the proposed research going hand-in-hand with the efficiency of $\text{CH}_4$ generation is the capture of $\text{CO}_2$, which by itself represents an important achievement.
Potentially, not only atmospheric CO₂ levels could be decreased, but the CO₂ emission could be converted into a powerful energy source as methane is. The realization of a complete “green circle” could be achieved by converting CO₂ emissions to biogas via biological means.

### 3.3 Central Activities

Central areas of activity and specific experimental activities are represented below in a graphical scheme which is connected to the project description reported in section 3.4.

![Figure 3. Schematic representation of the areas of activities, main and indirect outcomes related to each activity. Activities are described in higher detail in section 3.4.](image)

### 3.4 Description of the Project

#### 3.4.1 Coupling of Microbial Electrolysis Cell to Anaerobic Digestion to improve methane production

As it has been previously shown, hydrogen gas (H₂) can be produced in the anaerobic cathode compartment of MECs by applying a small external voltage in addition to the one generated by bacteria oxidizing organic material in the anode compartment [6,8,23]. The use of microbial biocathodes has been proven to facilitate the process of H₂ formation by catalyzing the electron transfer from the cathode to H⁺ ions through extracellular electron transfer mechanisms [9]. The produced H₂ can be used as substrate of reaction (1) shown in section 1.1, leading to the formation of methane [6,14].

The proposed project aims at testing a set-up consisting of a MEC directly connected to a lab-scale anaerobic digester, having the anaerobic sludge continuously circulating between the two systems (Fig. 3). The formation of CH₄ and H₂ evolution will be monitored via gas chromatography and the performances of the system will be evaluated at different applied potentials, as different potential values can greatly influence the electron recovery towards the formation of H₂ or CH₄.
The system will be run at first using mixed microbial population from industrial anaerobic digesters and synthetic waste-water. The use of synthetic waste-water of known composition will allow quantifying the degradation and conversion of organic and inorganic compounds both in the anaerobic digester and in the MEC. Furthermore, it will be possible to define whether and in which terms the same microbial consortium will have a different metabolic behavior in the anaerobic digester and in the MEC, respectively, by analyzing the composition of residual sources and metabolic products. This kind of information might be very useful in terms of wastewater treatment processes, as certain compounds can be degraded to different extent depending on the environmental conditions. Therefore, coupling a MEC to an anaerobic digester might add further value also in terms of degradation and/or conversion of compounds that are not currently eliminated via the anaerobic digestion process, such as excess of organic compounds (e.g. acetate) or sulphide [5].

Furthermore, as CH₄ synthesis derives from CO₂ reduction, it is important that CO₂ levels are not limiting in the system, in order not to impair CH₄ yields. Therefore, an additional advantage of such system is the possibility to capture CO₂ generated from other processes. In particular, as one of the main research focuses of the Industrial Biotechnology group is production of bioethanol through fermentation of renewable organic materials, an additional test will be performed to define the efficiency of CO₂ capture from fermentation off-gas and its conversion into CH₄. The feasibility of such approach would be a proof of concept for “highly integrated green technologies” towards the generation of fuels.

Throughout the process the following parameters will be monitored and measured:

- Cell voltage and current generated by MEC
- CH₄, H₂ and CO₂ (via gas chromatography)
- Levels of organic compounds in waste water influent and effluent (via HPLC)
- Levels of nitrate, nitrite, sulphate, sulphide and phosphate

All the measurement reported above will be carried out at the level of the different components of the system (Fig. 3) and at different applied potentials. Influenst and effluents will be characterized at
consecutive time points, in order to define which factors are mainly influencing the performance of the system and the microbial metabolism.

3.4.2 Characterization of microbial populations

The outcomes of such analyses, in connection with the measurements listed in section 3.4.1, will provide a better understanding of the relationships between environmental changes in the system and changes in the microbial populations. This knowledge will help in defining possible changes that might be brought to the system in order to favor or thwart certain species according to their influence on the performance of the entire system.

Microbial populations that will be used as biocatalyst in the MEC will be the endogenous ones present in the waste and reject water samples retrieved from wastewater treatment plants. The microbial consortia will be characterized by identification of the species thriving in the anaerobic digester and in the MEC using molecular biology tools based on sequencing species-specific genes. Although, typically, only bacterial species would be taken into consideration, in this project we will also include the possibility to identify eukaryotic microbial species, such as yeasts and other fungi, based on results of a preliminary study showing their presence in MFC fueled with waste-water.

Microbial consortia are typically subjected to specific dynamics due to several environmental factors. In order to follow and understand the dynamics of the microbial populations thriving in the described system, population analysis will be performed in the original wastewaters and at different time points in the MEC chambers and in the anaerobic digester. Population dynamics specifically related to differences in the applied potential will also be considered.

Techniques to be used for characterization of microbial consortia:

- Extraction of DNA from complex samples and from biofilm
- Amplification and sequencing of 16S rRNA (for bacteria and archaea) and 18S rRNA (for eukaryotes) genes
- Classical isolation techniques for different bacterial species

3.4.3 Use of metal foam electrodes in bioelectrochemical systems

Thanks to a collaboration established with the group of Materials for Mechanical Applications, Dept. of Mechanics at Politecnico di Milano (Milan, Italy), we have already carried out an exploratory study on the potential use of foam metal electrodes as electrodes in MFC systems (see section 1.3). The study demonstrated that the peculiar structure of the foam electrodes is advantageous for MFC performances, as using stainless steel foam electrodes resulted in a neat increase of the current generated in MFC systems compared to the use of massive electrodes.

The peculiar foam-structure, that we demonstrated improving the MFC performance, will have potential advantage also for the MEC biocathodes, supplying higher specific surface for the formation of the microbial biofilm. This would potentially increase the efficiency of the biocatalysed external electron transfer from the cathode to CO₂.

Metal foam electrodes of different composition and structure (i.e. pore size) will be tested also within this project and will be compared to the performance of graphite electrodes (i.e. the typical ones used in MFC and MEC studies) in the same system configuration. Such comparative study will define the potential advantages given by the use of metals (e.g. higher conductivity) and also the influence that different composition of metals can have on the performance of the MEC. A very interesting property one would like to have in a MEC system is the possibility to use metals with good catalytic potentials for H₂ evolution but cheaper than platinum.
Therefore, different kinds of metal alloys will be tested according to their availability as listed below:

- Steel (i.e. carbon steel; stainless steels alloyed also by high content of Cr and Mo)
- Magnesium alloys
- Pig Iron (also alloyed pig iron with Cr)
- Aluminum alloys
- Brass
- Pb-S alloy

### 3.4.4 Implementation of the AD-MEC system using relevant waste-water and anaerobic sludge

After a full characterization of the described system, actual waste-water will be used as “fuel” for the anaerobic digester and the MEC, considering as starting point the optimal set-up conditions defined by the studies carried on with synthetic waste-water.

### 4. EXPECTED OUTCOMES

The research project proposed is expected to both **characterize the critical aspects** that define the effectiveness of an AD-MEC coupled system and test different setups aiming at improving the system.

The scientific knowledge gained through such study will **provide Göteborg Energi with a future possible technological application which could improve the efficiency of biogas producing plants.** Concomitantly, the described project will contribute to establish a **new scientific collaborative connection within Chalmers University of Technology** between the Industrial Biotechnology group and the Department of Civil and Environmental Engineering, by building an **interdisciplinary link** from which both collaborative partners will gain further knowledge in the field of bio-electrochemical systems.

The achieved deeper knowledge will contribute to find possible novel applications of BES in connection with further possible improvement of existing industrial processes (e.g. wastewater treatment, recycling of chemicals).

Overall, as anaerobic digestion for biogas production is attracting continuously growing interest, improving the efficiency of the process will certainly contribute to make a **step further in developing sustainable means for energy production.**

### 5. MILESTONES and TIME PLAN

#### 5.1 Milestones

**M1.** Effect of BES on AD performance: how externally applied voltage can stimulate the biogas production

**M2.** Identification of conversion processes occurring at anode and cathode, including oxidation of organics, formation of CH₄ at the cathode and conversion of inorganic compounds (e.g. sulfide).

**M3.** Definition of the performance of metal foam electrodes

**M4.** Definition of microbial dynamics in the BES and in the AD, depending on time point and applied voltage

**M5.** Definition of AD-BES setup using real waste-water
6. PARTIES PARTICIPATING and POTENTIALLY INTERESTED in the PROJECT

**Collaborators**

- **Oskar Modin, PhD**: Department of Civil and Environmental Engineering, Chalmers University of Technology, Göteborg, Sweden  
- **Carlo Mapelli, Prof.**: Materials for mechanical Applications, Dept. of Mechanical Engineering, Politecnico di Milano, Milan, Italy  
- **Industrial Biotechnology group**, Department of Chemical and Biological Engineering, Chalmers University of Technology, Göteborg, Sweden

**Organisations possibly influenced by the project or possibly influencing the project**

- **Göteborg Energi**: research department for Biogas production. Mr. Eric Zinn is our reference.  
- **Gryaab, AB**: municipal wastewater treatment plant, which has been already collaborating within the project 09-13 financed by Göteborg Energi Forskningsstiftelsen.  
- **Chalmers Energy Initiative**: consortium of research groups and companies established at Chalmers University of Technology based on the raised level of funding for strategically important research areas appointed by the Swedish Government in the Bill “A Boost to Research and Innovation”. The industrial Biotechnology group is part of the research area “Energy Combine”

7. REFERENCES