Bioelectrochemical biogas upgrade: A novel technology for reduction of carbon dioxide (CO_2) into methane

I.A. Vasiliadou[1], A. Kalogiannis[1], A. Spyridonidis[1], A. Katsaounis[2], K. Stamatelatou[1]

[1]Department of Environmental Engineering, Democritus University of Thrace, 67100 Xanthi, Greece

[2]Department of Chemical Engineering, University of Patras, 26504 Patras, Greece

Purpose

Residual biomass, comprise a renewable energy source, that can be recovered into biogas through anaerobic digestion. Biogas upgrade, may lead to pure biomethane production, which can be used as vehicle fuel and natural gas substitute. Biogas upgrade is currently conducted using energy-consuming physicochemical processes or external H_2 source for biological CO_2 conversion to CH_4. Bio-electrochemical biogas upgrade, is an innovative method, in which CO_2 is converted into CH_4 by methanogenic microorganisms, in the bio-cathode of a Microbial Electrochemical Cell (MEC), using electric current as energy source, while no H_2 supply is required. The goal of the present study, was the development of a MEC for efficient biogas upgrade, with a biocathode of high electromethanogenic activity (Figure 1).

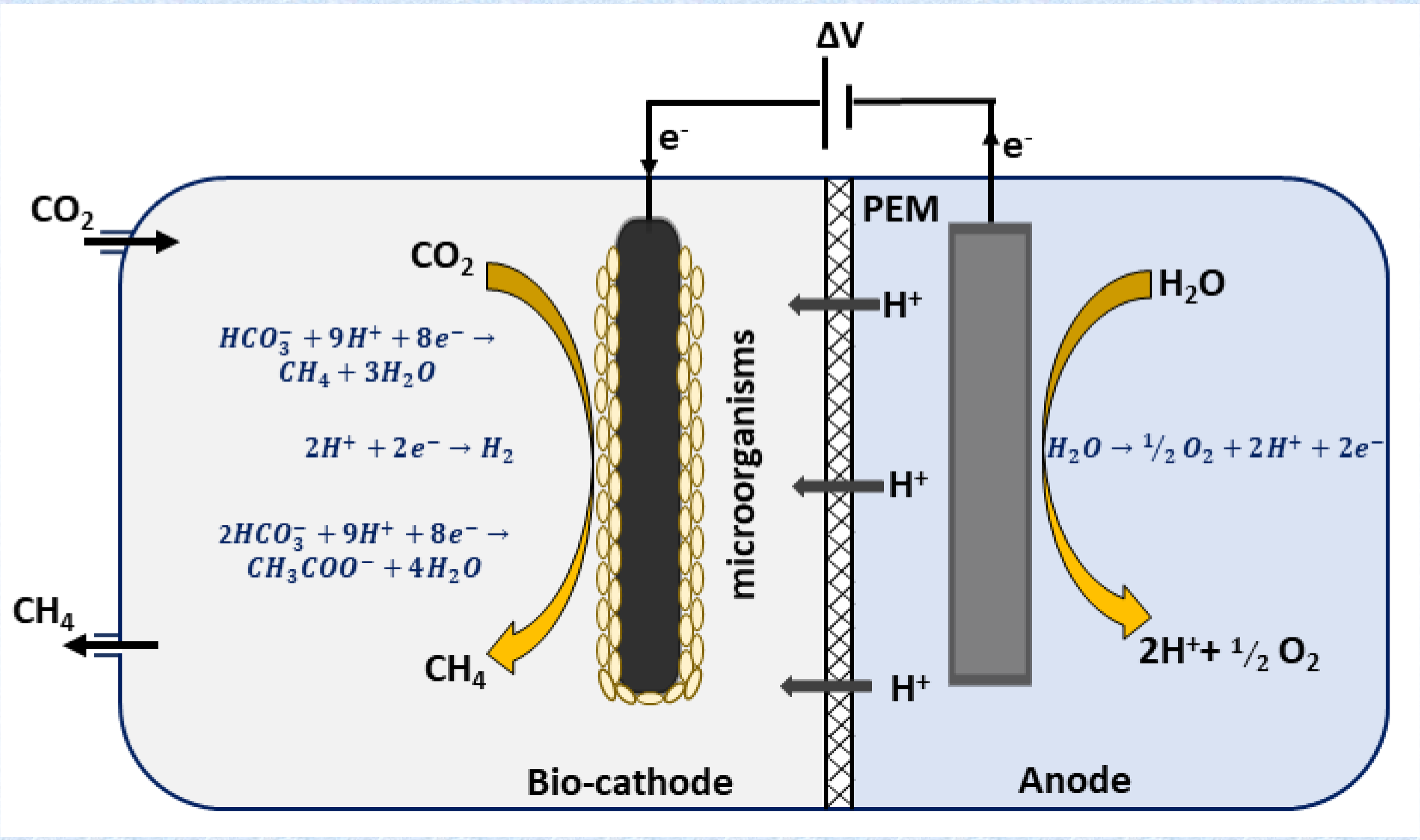

Figure 1. Bioelectrochemical biogas upgrade in a MEC.

Methods

A special designed H-type MEC (Figure 2), consisting of a cathode and an anode chamber (separated by a proton exchange membrane, Nafion117), has been used. The cathode chamber, was inoculated with a mixed culture of methanogenic microorganisms previously acclimated under biogas bio-upgrade conditions. The H-MEC, operated in a three-electrode configuration (working: carbon rod, counter: Pt/Ti, reference Ag/AgCl) with a potentiostat, which poised the bio-cathode at -0.7 V vs. standard hydrogen electrode (SHE) and monitored the current demand. Every 6 days, the catholyte and anolyte of the MEC were supplied with N_2/CO_2 gas mixture and N_2 gas, respectively. The conversion of CO_2 to CH_4 was evaluated periodically.

Cyclic voltammetry (CV) runs were performed, in the range of -0.9 to 1.2 V (scanning rate 5 mV/s) in order to determine whether the methanogenic culture was adapted to the electrochemical conditions.

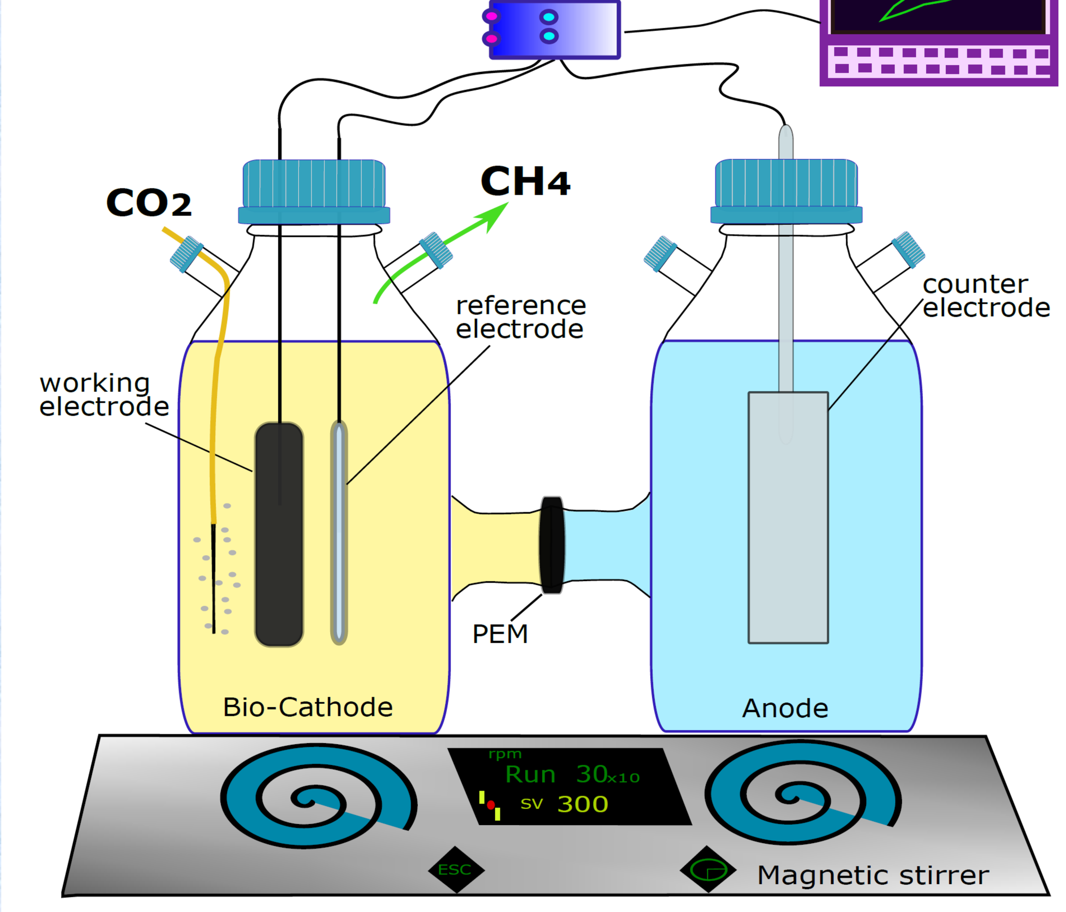

Figure 2. Microbial Electrochemical Cell set-up

Results & Discussion

Based on cyclic voltammetry runs, a highly electroactive methanogenic biocathode was developed in a short-time of period (Figure 3). Results suggested that after 14 days of acclimation (start-up period), microorganisms were interacting with the carbon-electrode, and thus a greater catalytic wave, related to hydrogen production, appeared (started at a potential of -0.7 V), as compared to the corresponding CV of the initial inoculum. In addition, a reductive peak allocated at -0.15 V, appeared and related to methanogens activity.

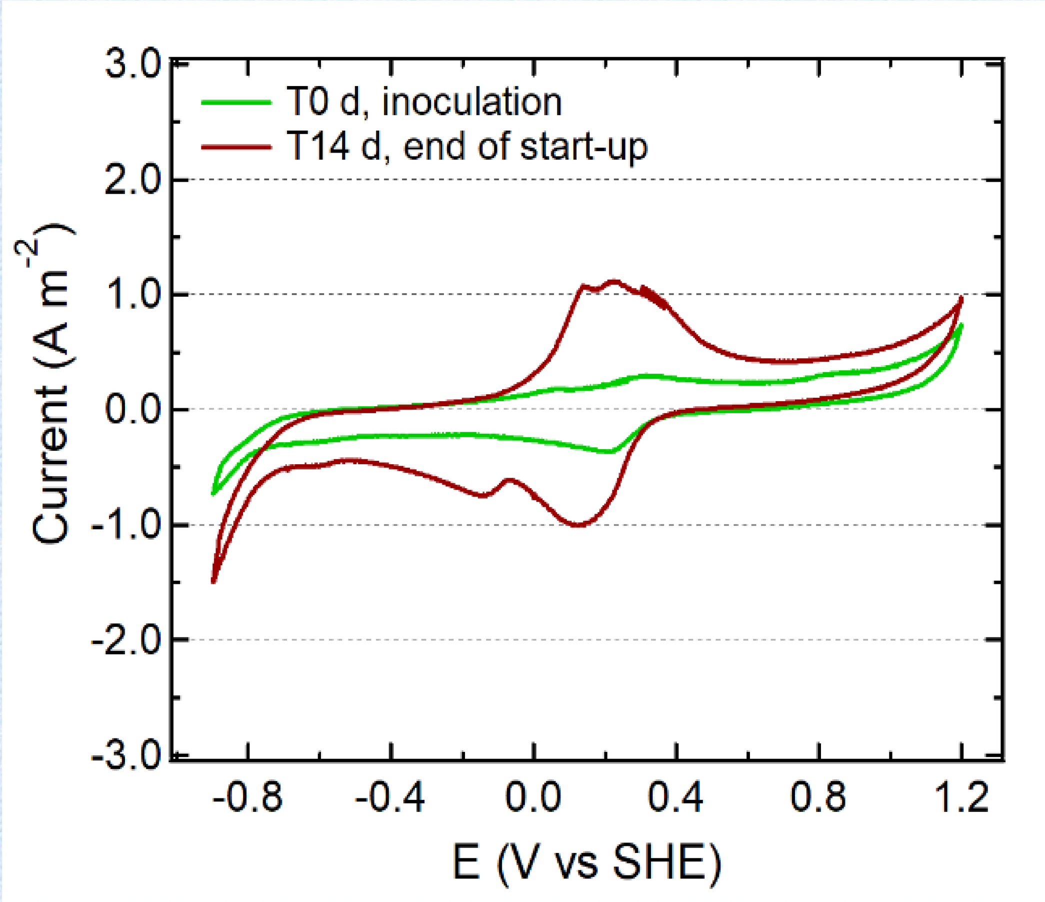

Figure 3. CVs at the beginning (inoculation, Time 0d) and at the end of the start-up period (attached microorganisms, Time 14d) under N_2/CO_2 saturated conditions.

Figure 4 shows the current density vs. time for the H-MEC and the methane and hydrogen production rates, during the operating period. Results suggested, that at cathode potential -0.7 V, microorganisms were able to drive the reduction of CO_2 to CH_4 mostly via direct electron transfer; however, indirect pathways by using the bioelectrochemically produced hydrogen and acetate, could also be occurred. The system's methane production rate (MPR) was 42.8 mmol m^{-2} d^{-1}, with a high electron capture efficiency (Coulombic efficiency, CE 95.4 %) (Figure 4).

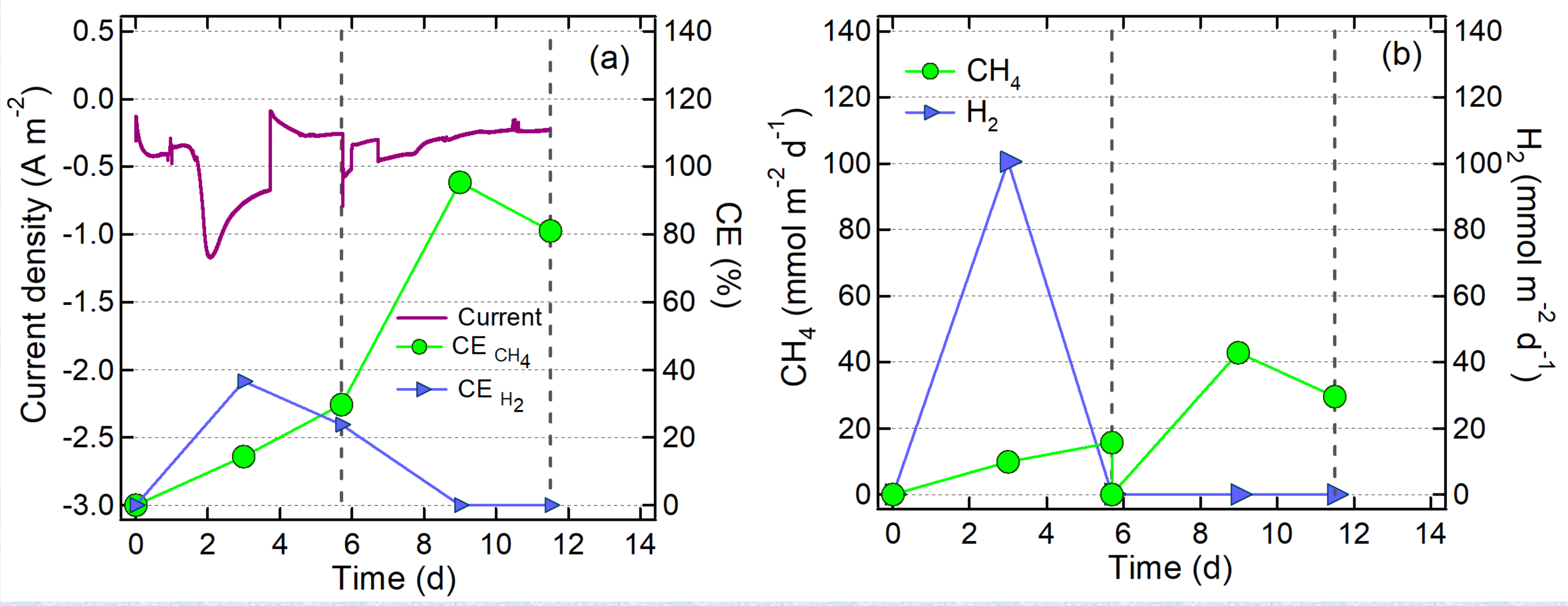

Figure 4. Bioelectrochemical experiment at -0.7 V with the enriched electroactive methanogenic culture. (a) Time course of current and coulombic efficiency and (b) Methane and hydrogen production rates. Vertical lines indicate biocathode's resupply with N_2/CO_2.

Conclusions

This study showed the development of an electroactive methanogenic biocathode in a two-chamber MEC at a potential of -0.7 V, using as inoculum methanogenic microorganisms pre-acclimated under biogas bio-upgrade conditions. Results, showed that the presence of hydrogen and acetate in the bio-cathode, could enable the indirect bioelectrochemical methane production. The CO_2 resupply of the biocathode, increased the CH_4 production rate and enhanced the direct electron transfer for electromethanogenesis; however, indirect formation via acetate could also be occurred. The proposed system achieved considerable MPR, after a short start-up period.

Acknowledgments

This research is co-financed by Greece and the European Union (European Social Fund- ESF) through the Operational Programme «Human Resources Development, Education and Lifelong Learning 2014-2020» in the context of the project "Bioelectrochemical biogas upgrade into methane (CH_4): An innovative method of carbon dioxide (CO_2) conversion" (MIS 5050548)."

European Union
European Social Fund

Operational Programme
Human Resources Development, Education and Lifelong Learning
Co-financed by Greece and the European Union

ΕΣΠΑ 2014-2020
ανάπτυξη - εργασία - αλληλεγγύη