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Bioelectro-catalytic valorization of dark fermentation effluents by acetate oxidizing bacteria in bioelectrochemical system (BES)

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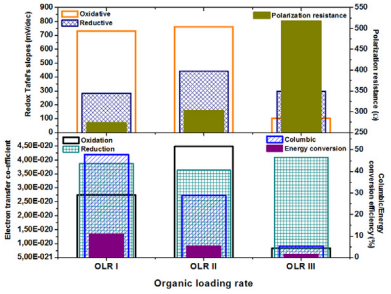
HIGHLIGHTS

- An MFC was operated with enriched biocatalyst from farm manure.
- Utilizing dark fermentation effluents as primary substrate in MFC.
- Bioelectrochemical analysis along with the wastewater treatment was studied.
- Shifts in organic acid contents during the process were linked to power output.
- Fraction of COD converted to power was calculated and compared with literature.

GRAPHICAL ABSTRACT



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ABSTRACT

Biovalorization of dark fermentation effluent (DFE) in a microbial fuel cell (MFC) was studied using the biocatalyst enriched from farm manure. The MFC performance was evaluated in terms of power density, substrate degradation, energy conversion efficiency and shifts in system redox state with operation time and organic loading rate (OLR). Higher power density of 165 mW m^{-2} (12.5 W m^{-3}) was observed at OLR I, which dropped to 86 mW m^{-2} at OLR II and 39 mW m^{-2} at OLR III. The substrate degradation was also higher at OLR I (72%) and diminished with increasing the OLR. The pH showed rapid drop and fluctuations initially when shifted to DFE but adapted over time. Higher coulombic efficiency was observed (48% at OLR I) contributing to a total energy conversion of 11%, which is higher compared to the available literature. However, the MFC performance declined at higher OLR with respect to all the performance indicators. DFE consisted of residual sugars from first stage process along with the volatile fatty acids (VFAs) and alcohols, which contributed for the generation of organic acids with their simultaneous consumption and led to VFA increment in spite of COD removal. Cyclic voltammograms along with the derived electro-kinetics supported the observed shifts.

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1. Introduction

The current energy crisis and the resulting price escalations have drawn considerable attention toward the exploration of alternative eco-friendly renewable energy sources [1]. The biological production of hydrogen from waste organics has attracted the global attention as a clean energy source and a potential alternative

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to fossil fuel energy. Biohydrogen production *via* dark fermentation (DF) has garnered significant focus among the available methods due to its low economics and ease of operation [2]. Biohydrogen production is the initial part of anaerobic digestion (AD) where the organic substrate is biologically converted to volatile fatty acids (VFAs), releasing H_2 as a by-product, through a series of biochemical reactions [2,3]. On the other side, there is a gap in net energy balance for this process due to the greater consumption of heat and electricity compared to that produced as hydrogen. Also, the left over oxidized metabolites, i.e. VFAs and alcohols, in the DF effluent has a high stored energy values and carbon residues which impede the treatment process of this effluent [2,4,5].

The trend of industrial biotechnology to treat wastes and residues for landfill disposal was switched to direct valorisation to valuable materials and energy, as a result of economic concerns and the increasing concern about low availability of raw materials [6]. Moreover, the high energy consumed by traditional wastewater treatment technologies drive the demand for alternative and effective systems that can also fulfil the energy gap, by its ability to produce clean and sustainable energy while assimilation of wastewater simultaneously [7,8].

In these circumstances, development of processes for the effective utilization of wastewater to generate sustainable energy gained attention of the research fraternity across the globe. Microbial fuel cell (MFC) is one of these technologies that can transform the substrate chemical energy into electrical one by the metabolic activity of microorganisms [9–11]. The main redox reactions of MFC include the oxidation of substrate by anodic microorganisms to produce electrons and protons, in which electrons are transferred to the cathode through the external circuit, while protons are exchanged through the proton exchange membrane (PEM) separating the anode and cathode compartments [10,12]. A wide range of wastewaters have been utilized as anodic fuels in MFC for the power generation as well as their valorization [13]. However, the coulombic and energy conversion efficiencies are lower in these systems due to different electron losses [14–16]. In this context, some researchers started working on integration of multiple bioenergy systems for various forms of energy generation, which is considered more viable than single bioenergy system in terms of energy losses and substrate utilization. In this direction, secondary integration to acidogenic dark fermentation process gained more interest because the fermentation process simplify the complex substrates into simple volatile acids which can be further converted to various value-added products. The short chain fatty acids are energy rich flexible central commodities of bacterial metabolism and can be converted to diverse value added products by altering the bioprocess [17,18]. Integration of photo fermentation for H_2 production [4,19], microalgae growth for lipid production [20], microaerophilic fermentation for bioplastics production [21] and MFC for power production [22–26], are some of the known examples. However, the photo fermentation and MFC operation for the utilization of acid rich effluents though studied well in the past has not yet been fully exploited, especially in the directions of energy conversion efficiencies.

In the present study, an attempt was made to utilize the DFE from sucrose fermentation process as substrate for power production in an MFC at increasing OLR. A detailed evaluation of bioelectrochemical and energy conversion parameters as well as wastewater treatment efficiency was carried out. Comparative evaluation of the results with the existing literature was also done.

Table 1
The characteristics and composition of the original wastewater acquired after dark fermentation (DFE).

Characteristics of wastewater	
pH	4.3
COD (mg l^{-1})	16220
Composition of wastewater[a]	
Sucrose	2687.18
Glucose	951.62
Fructose	4566.78
Xylose	26.36
Acetic acid	1862.29
Propionic acid	64.60
Butyric acid	1754.26
Isobutyric acid	43.94
Caproic acid	34.7
Lactic acid	752.3
Ethanol	1554.8
n-Butanol	2.6

[a] All the concentrations are in mg l^{-1}.

2. Materials and methods

2.1. DFE characteristics

DFE was collected from a continuously running dark fermentation bioreactor at VITO, Belgium, for the production of hydrogen using sucrose as the main substrate. The bioreactor had been running for 150 days at various organic loading rates and retention times. When an effluent sample was taken for the MFC tests, the hydraulic retention time (HRT) was 4 h and the OLR was 96 g COD/L d. The chemical oxygen demand (COD) of the collected DFE was 16.2 g L^{-1} which was stored at 4 °C and brought to room temperature prior to use. The collected wastewater had a pH value of 4.3. The composition of the DFE and it's characteristics are represented in Table 1.

2.2. Biocatalyst

The cattle manure was collected from a livestock farm in Boeretang area in Mol, Belgium, and was used as the raw source of inoculum. The manure (100 g) was dissolved in 1 L of PBS under continuous stirring on a magnetic stirrer (500 rpm) for about 4–5 h, with intermittent mixing by a glass rod to avoid the settlement of sludge particles. The sludge was filtered through large porous metal sieve, so that all the larger particles including stones, threads, fibres, grass, etc., were separated and the fine sludge was collected, after which it was mixed with 40 mM sodium acetate and enriched for 48 h (400 rpm; room temperature). The sludge was then re-filtered with small porous metal mesh to collect the finest part of sludge which includes the biocatalyst and was used as inoculum.

2.3. MFC construction

Dual chambered MFC was fabricated using polyvinylidene difluoride (PVDF) material, each chamber with working volume of 0.03 L and operated in sequential batch mode (Fig. 1). The two compartments were separated by ion permeable dense separator consisted of Zirfon®. Carbon cloth (Mast Carbon™, UK) and Vito CORE™ carbon electrodes (both with a projected surface area of 10 cm^2), were used as anode and cathode, respectively. Stainless steel mesh was used as a current collector for anode. Prior to use, the separator and electrodes were pre-treated in PBS as previously described [27]. Ag/AgCl–3 M KCl (+197 mV vs. SHE) was used as reference electrode (Radiometer Analytical, France). The ion permeable separator (Zirfon®) was placed between the working electrode and the counter electrode, in order to prevent interference of gases (O_2 or H_2) which can be produced at the counter electrode during the polarization measurements. Ambient air (21% O_2) was used as catholyte at an overpressure of 5 m bar (g). The DFE was used as the anolyte by diluting in buffer (mg L^{-1}: NH_4Cl (200),

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