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COMBINED ANAEROBIC - AEROBIC BIOELECTROCHEMICAL SYSTEM FOR AMMONIUM, SULFIDE AND CARBON REMOVAL FROM ETHANOL STILLAGE

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ABSTRACT

The organic matter (as an electron donor) could be oxidized at an anode in the bioelectrochemical systems (BESs), to produce electrons and protons with the biocatalysts of electrochemically active microorganisms. Electron acceptors at the cathode could accept them. By combining an anaerobic digestion reactor with BES, the chemical composition of biogas can be improved - the yield of methane can be increased at the expense of carbon dioxide. If an aerobic reactor is included in this combined system, complete decomposition of organic matter, generation of energy (biomethane) and nitrogen removal is achieved.

The efficiency of carbon, sulfide and ammonium ions removal during anaerobic digestion of ethanol stillage in an integrated BES system was studied in 5 modes of operation - without BES, microbial fuel cell (MFC) and microbial electrolysis cell (MEC at 0.6, 0.8 and 1.0 V external voltage). In MEC mode, the gas composition is improved (increased amount of methane to 74 %, vol.), and the content of sulfates and hydrogen sulfide is reduced. At 1.0 V external voltage, 94.1 % removal of organic matter and 90.91 % removal of NH_4^+ are achieved.

Keywords: BES, ethanol stillage, COD, hydrogen sulfide, ammonium, biomethane.

INTRODUCTION

The production of renewable biomass often involves the generation of co-products, by-products, or wastes. Anaerobic digestion (AD) has several advantages over aerobic treatment processes including renewable energy generation (biomethane), reduced energy costs through the elimination of aeration, and reduced sludge treatment and disposal expenses. The aerobic treatment usually depends on the oxidative activities of microorganisms. In the recent century, AD has been used for bioenergy generation from organic solid and liquid wastes. Bacterial respiration can be utilized to generate power in a fuel cell when combined with an external circuit. This creates an opportunity to induce biofilm growth, and thus electricity, from bacteria using controlled potential or electrical voltage. The advantage of biomethane from AD is that it can easily be stored or transported. Compression, transport in pipes, and storage involve mature technologies and could rapidly be integrated into existing infrastructure [1, 2].

Bioelectrochemical systems (BESs) are promising new technology. They contain an anode and cathode with or without a membrane. The BESs convert organic matter into protons and recoverable electrons in an anode compartment. Electrons pass through an external circuit to the cathode and conduct reduction reactions. Hydroxide ions are produced by the reduction of oxygen at the cathode. The BES systems demonstrated a high potential for organic wastewater treatment. The electroactive bacteria (EAB) can remove the organic carbon easily from wastewater while producing biological renewable energy in the form of electricity in a microbial fuel cell (MFC) - a particular type of BES. Two systems are most commonly used for wastewater treatment: a microbial fuel cell or a microbial electrolysis cell (MEC). Anodic reactions are almost the same in both configurations. By a load in MFC Anode and cathode are connected, while in MEC anode potential is regulated by an external voltage [2 - 4].

The use of a hybrid AD-MEC system has been proven to increase the quality of the obtained biogas by reducing CO_2 to CH_4, which can reduce the cost of biogas purification [5]. An anaerobic digestion and anodic oxidation from a MEC were combined to improve the wastewater degradation process. Methane from microbial electrolysis cells has been suggested as an energy-friendly effluent polishing step for digester effluents, most likely entailing low sludge production rates and no aeration costs [6, 7].

AD-BES systems are also used for organic carbon (Chemical Oxygen Demand), sulfates, sulfides, nitrates, and heavy metals removal [8, 9]. The not consumed chemical oxygen demand (COD) for electricity production is degraded by competitive metabolisms that use other electrons and COD for growth and reproduction. There is a positive relation between COD value and voltage generation in MFCs, due to the oxidation of organic matter, because this increases the electron transport and consequently the power density. Also the recovery of other products such as ammonia increases the MFC viability in wastewater treatment [9 - 12]. Heavy metals (Cr and Ag) and other by-products (CH_4 and H_2) can be recovered by MFC, however, specific conditions like microorganism growth, anode and cathode media, and concentration must be considered to ensure a high-power density production. Nitrogen in the form of ammonium ions can be removed from wastewater by ammonification, nitrification, and denitrification. The microorganisms perform the removal of NH_4^+ from wastewater. The ions are transported then to the cathode chamber. Ammonium transport from anode to cathode is carried out by diffusion and migration. Moreover, supplementary aeration increases the ammonium removal to nitrate that is used as an electron acceptor in the respiration of anaerobic denitrifies [9].

Rahmani et al. investigated ammonium removal and its recovery with parallel power generation from wastewater in BES. They built a two-chamber Plexiglas BES with a volume of 450 mL, which was divided by a cation exchange membrane (Nafion, 117) with the establishment of two carbon plates (50 mm x 40 mm x 3 mm) as anode and cathode electrodes [13]. From the results can be concluded that BES can be an effective process for removing high concentrations of organic matter and ammonium from industrial wastewater. It was obtained the maximum efficiency with 150 mg L^{-1} input ammonium concentration and 10 g L^{-1} COD concentration and 0.368 V external voltage (94 % for the NH_4^+-N removal) and with a 2 g L^{-1} COD (78 % for maximum organic matter removal) and 0.481 V external voltage.

In the present study, a combined BES system with an aerobic and anaerobic reactor was constructed. The process of utilization of an ethanol stillage under different BES modes- without BES, as a microbial fuel cell, and as a microbial electrolysis cell (with cation exchange membrane) with 3 different external voltages (0.6, 0.8, and 1.0 V) was monitored. The main goal of the research is to establish the possibility of using BES in combination with anaerobic and aerobic bioreactors, with the aim of a higher degree of organic substrate utilization, removal of sulfur and nitrogen compounds, and improvement of the biomethanation process.

EXPERIMENTAL

Substrate and inoculum

Ethanol stillage was obtained from a winery in Svetovrachene village, Bulgaria. After obtaining the ethanol stillage, it was stored in a cool place at 4°C. Before using it, the wastewater was neutralized to pH 7.5 with NaOH and diluted 4 times. The aerobic and anaerobic activated sludge was taken from "Almagest" AD, Verinsko village, Bulgaria.

Reactor and BES design

The experiments were carried out in a laboratory installation of bioelectrochemical system, in which the possibility of working in two modes is foreseen - as microbial fuel cell and microbial electrolysis cell (Fig. 1). For this purpose, a two-chamber construction of the BES "sandwich" type with a separator was used, separating the cathode from the anode zone by a cation exchange membrane (CEM) type - CMI-7000S, Membrane International Inc. Before the start of the experiments, the membrane was treated with a solution of 0.5 M NaCl at 25°C for 72 h. Graphite plates with dimensions of 100 x 100 x 6 mm and a geometric area of 0.021 m^2 were used for electrodes in the BES. The area of the CEM was 0.01 m^2. The volume of the cathode and anode sections are the same - 100 mL. Before each series of experiments, the membrane and electrodes were replaced with new ones.

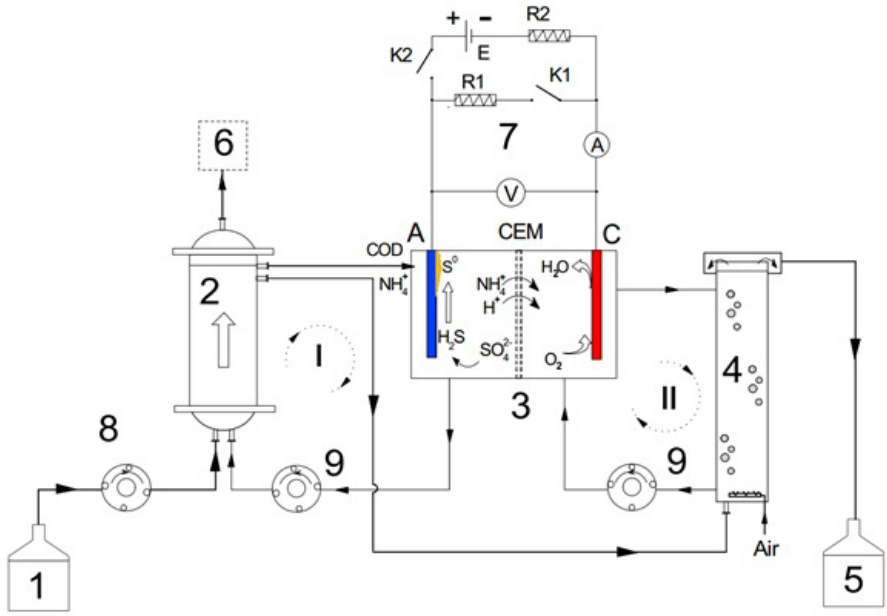

Fig. 1. Combined BES system with aerobic and anaerobic bioreactor: 1 - substrate input, 2 - Upflow Anaerobic Sludge Blanket (UASB) bioreactor, 3 - BES with cation-exchange membrane (CEM), 4 - aerobic bioreactor, 5- reactor outlet, 6 - biogas, 7 - load chain of BES, 8 - feed pump to the substrate, 9 - recirculation flow.

The anaerobic reactor was made of stainless steel with a working volume of 3 dm^3. The aerobic reactor was made of plexiglass with a 1.5 dm^3 working volume and an aeration time of 1 dm^3 min^{-1}. Wastewaters enter the anaerobic reactor, where oxidation of organic matter by a methanogenic consortium takes place, and with the help of a recirculation pump passes through the cell anode. The flow from the anaerobic reactor is fed to an aerobic reactor for aerobic nitrification using another pump, where the ammonium ions are converted to nitrate. A pump through the cathode zone of the cell where nitrate reduction takes place recirculates the aerobic reactor stream. Electrons produced at the anode are transferred to the cathode through an external resistor, and protons diffuse to the cathode through the CEM along with ammonium ions. Biogas was collected in a gas bag, which was attached to the top of the anaerobic reactor. The contact time in the anaerobic reactor was 11 days, and in the aerobic one - 6 days. Once a day, 273 mL of wastewater was fed to the anaerobic reactor.

BES operations

Studies have been carried out in 5 variants of operation of the laboratory installation. Initially, according to the technological scheme (Fig. 1), a mode of operation was investigated, without turning on the BES, following the anaerobic biomethanation reactor (UASB), an aerobic bioreactor was connected in series. In the other 4 options in the technological scheme, the anode and cathode zones of the BES were connected in parallel, respectively to the anaerobic and aerobic reactors, with the liquid phases being recirculated continuously by peristaltic pumps (9) (with a flow rate of 5 dm^3 h^{-1}) in recirculation loops I and II (Fig. 1). The bioelectrochemical system has been successively investigated in 4 variants - as a microbial fuel cell (MFC) with an external load resistance of 100 Ω and as a microbial electrolysis cell (MEC) with applied 3 external voltages - 0.6 V, 0.8 V and 1.0 V. For all 5 modes a constant flow rate of the feed pump (8) is provided for a period of 11 days and providing the above-mentioned contact times in the two bioreactors.

For a shorter and easier description of the results obtained in the different modes of operation of BES, the following notations are introduced: *MFC* or *AD-MFC* for microbial fuel cell mode; $MEC_{0.6V}$, $MEC_{0.8V}$, $MEC_{1.0V}$ or *AD-MEC 0.6V, AD-MEC 0.8V, AD-MEC 1.0V* for microbial electrolysis cell mode and corresponding applied voltage; *without BES* - control experiment without including BES.

Experimental measurements and calculations

The sulfates concentrations were determined spectrophotometrically by $BaCl_2$ reagent at a wavelength of 420 nm and hydrogen sulfide in liquid - by using test 1-88/05.09 of "Nanocolor" at a wavelength of 620 nm. COD was measured with Hanna instruments reagents according to APHA (1992) [14]. Total Kjeldahl Nitrogen (TKN) and ammonium nitrogen (NH_4^+-N) were determined via standard methods by Keldahl apparatus - UDK 127, VELP SCIENTIFICA, Italy. Short-chain fatty acids and polysaccharides were analyzed by the HPLC system. The biogas composition was measured using a portable "Draeger X-am 7000" gas analyzer. Milli-gas counter MGC-1, Ritter, measured the biogas production.

The electrical parameters of the BES were measured with a Keithley 175 digital multimeter, and a precision potentiometer with a maximum value of 11 kΩ was used for the load resistance. The maximum value of the power and the current density was measured to establish polarization curves and the power curves in the mode of operation of BES as MFC. The current and power density is calculated based on the geometric area of the electrodes in the anode/cathode chambers and the voltage across the external resistance (R1 or R2). To provide an external voltage source, a stabilized adjustable rectifier type PS-3005D was used when operating the BES in MEC mode. In the mode of operation of BES as an MFC, a load resistance R1 = 100 Ω was applied, in which it was previously established that a stable operation of the fuel cell with an optimal value of the current density and the power. To investigate the performance of the BES as a MEC, an external load resistance R2 = 10 Ω was used, which is of a minimum value so as not to limit the current between the anode and the cathode in the BES. The energy density of the product (the amount of energy that can be extracted from a unit volume or mass of the fuel) is a good way to define the energy efficiency of a given process. The specific energy efficiency (energy density) was calculated by equation 1:

$$Energy\ density\ \left[\frac{KJ}{kg}\right] = \frac{V_{biogas} * C_{CH_4}}{24.1} * 16 * 55.5 \quad (1)$$

in which V_{biogas} is the volume of the produced biogas (L), C_{CH4} - methane content in biogas (%), 16 - methane mass in 1 mol (g), 55.5 - methane gross calorific value (MJ kg^{-1}) and 24.1 - the volume of 1 mol methane by 20 ºC (L).

RESULTS AND DISCUSSION

Characterization of ethanol stillage

Table 1 gives the main characteristics of the used ethanol stillage (neutralized and diluted four times). The composition is almost the same in the different BES operating modes that have been studied. The ethanol stillage has a high organic load (40 - 60 g L^{-1}) and it has been found that stillage with a COD over 100 inhibits stable digestion [15]. This may be overcome by dilution for the analysis diluted four times to make COD around 10 g L^{-1}.

Raffinose is a trisaccharide (galactose, glucose and fructose) from a family of soluble sucrose derivatives that have transported and energy carbon function in some plants. This carbohydrate is most commonly present in legumes and is also found in sugar beet molasses and whole grains. Raffinose can be fermented making short-chain fatty acids (acetic, propionic, butyric acids) [16]. The sulfates concentration was between 123 and 154 mg L^{-1}, TKN was 302 - 315 mg L^{-1}. The high content of protein (1.89 - 1.97 g L^{-1}), organic matter (9.21 - 10.00 g L^{-1}) and lactic acid (1.5 - 1.9 g L^{-1}) make the ethanol stillage a suitable substrate for methanation and bioenergy generation.

Electro-chemical characterization of the BES system

The polarization curve (Fig. 2) presents the maximum actual power generation and power density of the AD-MFC system (per total electrode surface area). According

Table 1. Main characteristics of ethanol stillage feeding the anaerobic reactor.

Parameter	Value
pH	7.5-7.6
COD, g L^{-1}	9.21-10.00
SO_4^{2-}, mg L^{-1}	123-154
Total Kjeldahl Nitrogen (TKN), mg L^{-1}	302-315
Protein, g L^{-1}	1.89-1.97
Dry matter, %	1.62-1.84
Raffinose, g L^{-1}	0.72-0.85
Lactic acid, g L^{-1}	1.5-1.9
Acetic acid, g L^{-1}	0.001-0.002
Butyric acid, g L^{-1}	0.11-0.47

to these data, the maximum values of current and power density are 133.6 mA m^{-2} and 50.1 W m^{-2}, respectively. These values were measured on the second day from the beginning of the experiment, and for a period of 11 days, they remained relatively stable. This is also confirmed by the timing diagram (Fig. 3) of the dynamics of the current density of the MFC, at an external load of 100 Ω. Fig. 3 shows the cyclic behavior of MFC and MEC during the biogas-producing process. This cyclicity is due to the fluctuations in electrical conductivity of the BES catholyte and anolyte, caused by the daily feeding of the system with the fresh organic substrate. The measured daily values of the current density, at four variants of BES, vary accordingly: MFC from 0.01 to 0.02 A m^{-2}, $MEC_{0.6V}$ from 0.4 to 1.1 A m^{-2}, $MEC_{0.8V}$ from 0.8 to 1.5 A m^{-2}, $MEC_{1.0V}$ from 1.5 to 2.9 A m^{-2}.

Energy generation (biomethane) by BES system

The graph in Fig. 4 shows that 8.6, 7.9, 7.96, 8.0 and 7.7 liters of biogas were produced in 11 days without BES, MFC, $MEC_{0.6V}$, $MEC_{0.8V}$ and $MEC_{1.0V}$, respectively. The composition of produced biogas is shown in Fig. 5.

In the case of the technological scheme without BES (anaerobic digestion only) 57 vol. % of the produced biogas is methane. As can be seen from the graph, there are significant amounts of CO_2 (24 vol. %), H_2S (0.034 vol. %) and H_2 (1 vol. %). In MFC mode, the methane content rises to 70 %, and in MEC mode, it is between 72 and 74 % at the different external voltages. In MEC or MFC mode, it can be seen that the increase in methane content in biogas is due to a decrease in carbon dioxide and hydrogen in it. Therefore, it can be concluded that in the MEC mode the composition of the produced gas

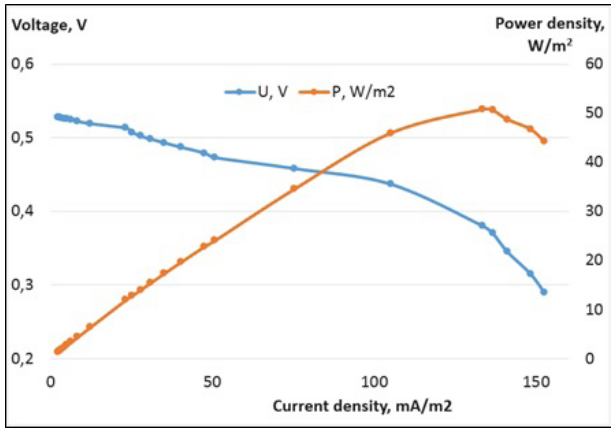

Fig. 2. Polarization curve and BES power curve in operating mode as MFC.

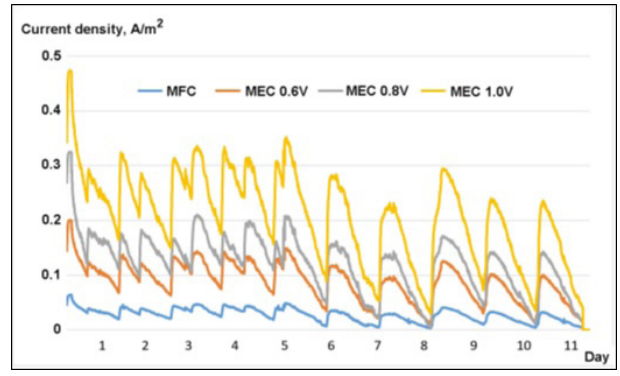

Fig. 3. Dynamics of the current density in different modes of BES operation - as MFC (100 W), MEC (0.6V), MEC (0.8V) and MEC (1.0V).

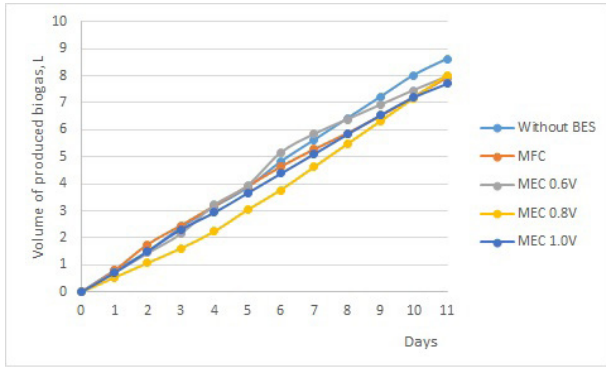

Fig. 4. Kinetics of biogas production from ethanol stillage in different operating modes of BES - without BES, MFC and MEC with different external voltage (0.6, 0.8 and 1.0 V).

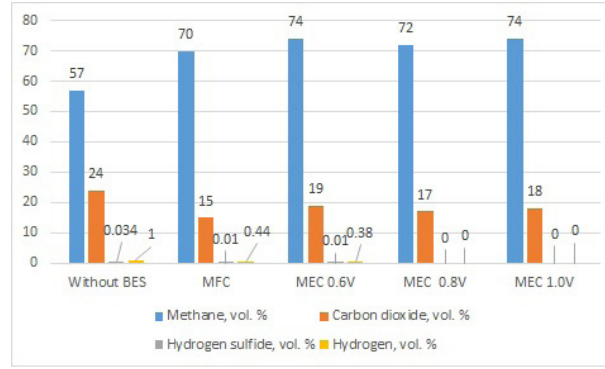

Fig. 5. Biogas composition by different operating modes of BES.

Table 2. Main parameters of output anaerobic reactor stream by 5 modes of BES.

Parameter	Without BES	MFC	MEC with 0.6V	MEC with 0.8V	MEC with 1.0V
SO_4^{2-}, mg L^{-1}	140.54	42.03	<1	<1	<1
$H_2S_{in\ liquid}$, mg L^{-1}	11.91	13.89	4.19	4.98	4.65
Raffinose, g L^{-1}	0.13	0.15	0.19	0.33	0.43
Lactic acid, g L^{-1}	0.29	0.27	0.14	0.23	0.24

Table 3. The COD and ammonium concentration in input, anode and cathode flow in the BES system by 5 different modes.

Parameter	Without BES	AD-MFC	AD-$MEC_{0.6V}$	AD-$MEC_{0.8V}$	AD-$MEC_{1.0V}$
COD input, g L^{-1}	9.21	9.21	10.00	10.00	10.00
COD anode, g L^{-1}	4.00	2.78	1.40	0.90	0.59
COD cathode, g L^{-1}	2.192	1.152	0.80	0.50	0.098
NH_4^+ anode, mg L^{-1}	166	155	139.1	125	108.9
NH_4^+ cathode, mg L^{-1}	77.06	48.5	45.5	30.5	9.9

and the stability of the process are improved compared to the MFC mode or without BES.

Ammonium, sulfide and COD removal from ethanol stillage by BES system

Sulfate-reducing bacteria (SRB) and methane-producing bacteria (MPB) always compete for the carbon source of anaerobic treatment processes to wastewater rich in sulfates and high organic content [17]. The production of toxic and corrosive H_2S has an inhibitory effect on these bacteria, while also degrading the composition of the resulting biogas [18]. In the present study, by using BES integrated into an anaerobic digestion reactor (AD), it is possible to realize one of the proven mechanisms for electron transfer in the anode zone - oxidation of H_2S on the anode surface to elemental sulfur (S^0) and other forms thereof [19].

As can be seen from the data in Table 2, in without BES mode there is minimal change in the sulfate content of the outlet stream (removal rate 8.74 %). After turning on the cell in the MFC mode, SO_4^{2-} content decreases by 72.71 %, and in the MEC mode, 100 % removal of sulfates from the ethanol stillage is achieved (at the three external voltages).

Dissolved hydrogen sulfide is in minimal amounts in MEC mode (4 - 5 mg L^{-1}) and slightly larger amounts without BES (11.91 mg L^{-1}) and MFC (13.89 mg L^{-1}) modes. This is the reason for the presence of hydrogen sulfide in the composition of the produced biogas in the last two. It is also evidence of incomplete oxidation of sulfides to elemental sulfur at the anode surface.

Raffinose and lactic acid decreased in value in all modes, indicating that there was a fermentation and degradation processes and they have been converted to methane. The best results for raffinose were achieved in MFC mode, followed by MEC under different external voltages and finally without BES. For lactic acid, the best results were obtained at $MEC_{0.6V}$, followed by $MEC_{0.8V}$, $MEV_{1.0V}$, MFC and without BES.

Table 3 shows the COD and NH_4^+ concentration in different flows (input to anaerobic reactor, in the anode and cathode zone) and Fig. 6 show the organic (as COD) and ammonium removal efficiency in BES system by 5 different modes - as MFC, MEC with 0.6, 0.8 and 1.0 V external voltage and without BES.

The organic matter efficiency removal was better in MFC and MEC mode vs without BES. As the external voltage increases, higher efficiency values are achieved - 91 % at $MEC_{0.8V}$ and 94.1 % at $MEC_{1.0V}$.

During the degradation of proteins, ammonia (NH_3) is released, which is very quickly reduced to free ammonium ions (NH_4^+). They are an important nutrient source and have an essential function in the process of methanogenesis. The process is inhibited

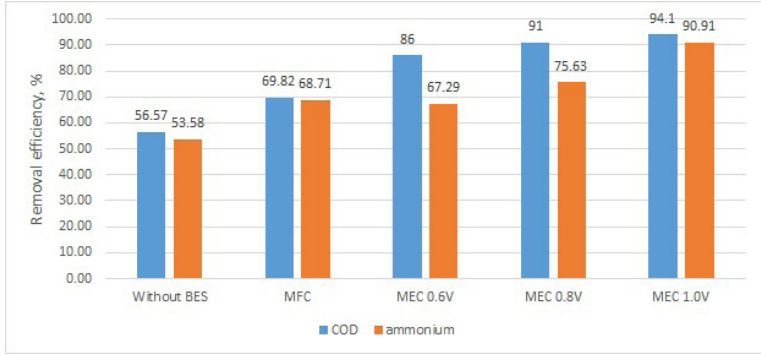

Fig. 6. The organic matter (as COD) and ammonium removal efficiency in the BES system by 5 different modes.

at high concentrations of NH_4^+. [9]. In the presence of a cation exchange membrane in the BES, the ions are migrated through CEM to the cathode zone. There, their concentration drops significantly, which is understandable considering the ongoing process of nitrification in the zone of the aerobic bioreactor. The best removal of ammonium ions was observed in MEC mode with 1.0 V external voltage (90.91 %), followed by $MEC_{0.8V}$. With $MEC_{0.6V}$ and MFC mode, the degree of removal is between 67 and 69 %, and in the mode without BES - only 53.58 %.

The obtained results for MEC mode at different external voltage correspond to those obtained by Lu et al., who reached 86 - 91 % removal efficiency of the chemical oxygen demand with a single-chamber MEC system [20]. They achieved these results when generating hydrogen using bovine serum albumin or peptone in a single-chamber MEC at 0.6 V external voltage. When the BES works as an MFC, the COD removal efficiency is 69.82 %. For comparison with a process without BES it was only 56.57 %. It obtained the maximum efficiency of organic matter by MEC with 1.0 V external voltage (94.1 %) and 90.91 % for ammonium removal efficiency.

Energy efficiency

The evaluation of the ethanol stillage energy potential biotransformation by different modes of the BES system is summarized in Table 4. The data shows that the energy potential in MEC mode is higher compared to MFC or without BES. At MEC with 0.6 V mode, the difference is 17 % compared to MFC and 18 % compared to anaerobic digestion only. The increase in energy potential with MFC is only 1 % compared to without BES.

Table 4. Estimated total energy content by different BES modes.

System	Estimated energy density, KJ kg^{-1}
Without BES	166.50
AD-MFC	168.50
$AD\text{-}MEC_{0.6V}$	203.35
$AD\text{-}MEC_{0.8V}$	190.24
$AD\text{-}MEC_{1.0V}$	196.65

CONCLUSIONS

In this study, the possibilities of using BES in combination with an anaerobic and aerobic bioreactor for the treatment of ethanol stillage have been analyzed to increase the degradation of organic substrate, remove sulfur and nitrogen compounds and improve the biomethanation process.

When comparing the results obtained in different BES operating modes, it was found that the best results were achieved in AD-MEC mode. When studying the effect of applied external voltage on the removal of organic and nitrogen compounds, the best results were obtained at 1.0 V (94 and 91 %, respectively), while no significant difference was observed in the removal of sulfur compounds. The composition of the produced biogas is better in all BES regimes compared to those without BES (from 57 vol. % to 70 - 74 vol. %). The results show that the MEC mode has greater advantages over MFC or without BES for the energy potential of the ethanol stillage biotransformation.

Based on the obtained results, it can be concluded that for the parallel generation of bioenergy and purification of ethanol stillage, an integrated system with MEC has an advantage over MFC.

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