



Enhanced Biogas Production from Tapioca Wastewater Through the Microbial Electrolysis Cell-Assisted Anaerobic Digestion Process at Various Urea Additions

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Abstract

In Indonesia, tapioca wastewater is one of the most abundant organic wastewater. It has a great deal of potential for use as a substrate for biogas, but it contains a high ratio of chemical oxygen demand (COD) to nitrogen (N). For this reason, adding nitrogen sources, such as urea, is crucial. Meanwhile, microbial electrolysis cell-assisted anaerobic digestion (MEC-AD) is a novel technology that can be applied to enhance biogas production. Thus, the purpose of this study was to ascertain how adding urea affected the biogas yield from tapioca wastewater through the MEC-AD process. There were six digesters, namely A (MEC-AD urea 0.25 g), B (MEC-AD urea 0.5 g), C (MEC-AD urea 1 g), D (MEC-AD urea 1.5 g), E (MEC-AD without urea), F (AD without urea). The MEC-AD process was carried out at room temperature using a batch system. The results revealed that the MEC-AD (without urea) generated a biogas yield 2.3-fold higher than AD alone (without urea). Then, an increase in urea addition in the MEC-AD process from 0 to 1.5 g enhanced biogas yield from 106.4 to 268.8 mL/g-COD. It means that in MEC-AD, the urea addition of 1.5 g generated 2.5 times more biogas yield than without urea addition. The MEC-AD with urea addition of 1.5 g had the most stable substrate pH and the highest volatile fatty acids during the process. The MEC-AD (without urea) gave a higher COD removal efficiency (21%) than AD alone (without urea), namely 14%. Then, an increase in urea addition from 0 to 1.5 g in MEC-AD increased COD removal from 21% to 38%. Therefore, the best variable was MEC-AD with a urea addition of 1.5 g. This innovation is expected to reduce environmental pollution and provide biogas as alternative energy to substitute the use of fossil fuels.

INTRODUCTION

Energy is a crucial resource for sustaining life, as it underpins all human activities (Budiyo et al., 2014). Resources for fossil fuels are depleting as a result of rising energy consumption brought on by population growth (Marendra, 2011). Therefore, the Indonesian government has a policy to replace fossil fuels with renewable energy sources. The government's focus on renewable energy is a strategic effort to secure Indonesia's energy future (Marendra, 2011).

Biogas, a sustainable and eco-friendly energy source, is generated through the breakdown

of organic materials by anaerobic microorganisms. This process not only provides renewable energy but also contributes to effective waste management (Khanal, 2008). In Indonesia, tapioca wastewater is a particularly abundant type of waste that can be utilized as a biogas feedstock.

Indonesia ranks as the second-largest producer of tapioca flour in Asia, surpassed only by Thailand, with 18.3 million tons expected in 2020 (Teguh et al., 2022). The manufacturing process of tapioca flour generates approximately 12 cubic meters (or 12,000 liters) of wastewater per ton of flour produced (Azizah et al., 2017). This wastewater originates from various stages,

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including washing, extraction, and settling. The wastewater from the tapioca flour industry not only emits unpleasant odors and causes water turbidity but also poses a significant pollution threat to aquatic environments if not adequately treated (Agung & Winata, 2017).

Generally, industrial organic wastewater is often directly discharged into water bodies without prior treatment and receives little attention from industry players, especially among small and medium-sized enterprises (Sugiharto, 1987). Tapioca wastewater, in liquid form, is directly discharged into river streams without undergoing any prior treatment, leading to environmental damage (Saputra et al., 2016). Meanwhile, solid waste is typically processed into materials with economic value. The solid wastes consist of cassava peels and "onggok" (cassava pulp). Cassava peels are utilized as animal feed, and cassava pulp is processed into tapioca flour (Indrianeu & Singkawijaya, 2019). As a result, effective and efficient waste treatment technologies are essential to treat this wastewater. One of them is anaerobic digestion. This technology can convert organic compounds of tapioca wastewater into biogas.

Tapioca wastewater contains high organic contents, including 21.067 % glucose, 18.9 % carbohydrates, and 51.040 % vitamin C (Rivera et al., 2019). According to (Sensih & Prayitno, 2020), this wastewater also contains high chemical oxygen demand (COD) in the range of 7,000 to 30,000 mg/L and biological oxygen demand (BOD) in the range of 3,000 to 6,000 mg/L. Hence, tapioca wastewater is a suitable raw material for biogas production because of these characteristics.

During anaerobic digestion (AD), the ratio between COD to nitrogen (COD:N) is one of the key factors (Syaichurrozi et al., 2013; 2016). The substrate with a high COD:N ratio is easily decomposed to volatile fatty acids (VFAs). Accumulation of VFAs can lower the pH and destabilize the fermentation process, thereby hindering methane production (Syaichurrozi, 2017). During AD, nitrogen sources can be degraded to ammonium or ammonia which can act as natural buffers, thus liquid pH will be stable. In addition, nitrogen (N) plays a crucial role in the formation of microbial cells that drive anaerobic processes (Cremones et al., 2021). Therefore, the amount of nitrogen available in the substrate directly impacts the efficiency of biogas production. Previous research (Yu et al., 2018) revealed that AD

of the substrate with too much carbon-to-nitrogen ratio generated a low biogas yield. Urea, a readily digestible nitrogen source for a variety of microorganisms, can be added to the substrate to increase nitrogen levels (Budiyono et al., 2013). The tapioca wastewater contains a high COD concentration and a low N concentration. Hence, urea addition to increasing the nitrogen level in tapioca wastewater is important to be conducted to succeed in the AD process.

Enhancing substrate breakdown and biogas production can be achieved by integrating the AD process with other methods. Combining anaerobic digestion (AD) with the microbial electrolysis cell (MEC) is an appealing idea. In the integration of microbial electrolysis cell and anaerobic digestion (MEC-AD), electrodes (anode and cathode) are incorporated directly into the anaerobic digestion reactor. This allows both MEC and AD processes to occur simultaneously within a single reactor, simplifying the setup. The MEC-AD technique has been previously utilized by Syaichurrozi et al. (2024) in the treatment of tofu wastewater with the addition of iron (Fe). The MEC-AD system successfully enhanced biogas production by 14.4-114.5% and improved pollutant removal efficiency by 1.09 to 1.63 times compared to the AD process. Research by Joicy et al. (2022) investigated the use of MEC-AD in treating activated sludge waste at different voltages (0.3-0.9 V) and found that it produced 27.2%-44.8% more methane than the AD alone.

Based on the information above, biogas production from tapioca wastewater can be enhanced through the MEC-AD process with urea addition. The current study is new because the application of MEC-AD technology for generating biogas from tapioca wastewater has not been explored. Thus, the current study aimed to evaluate the impact of urea addition on biogas generation from tapioca wastewater using MEC-AD technology.

MATERIALS AND METHOD

Materials

The substrates utilized in the current research included synthetic tapioca wastewater and cow rumen fluid as inoculum. The synthetic tapioca wastewater was made by dissolving 10 g of tapioca flour in 1 L of water (Budiyono et al., 2018). The tapioca wastewater contains COD 6419 mg-O₂/L

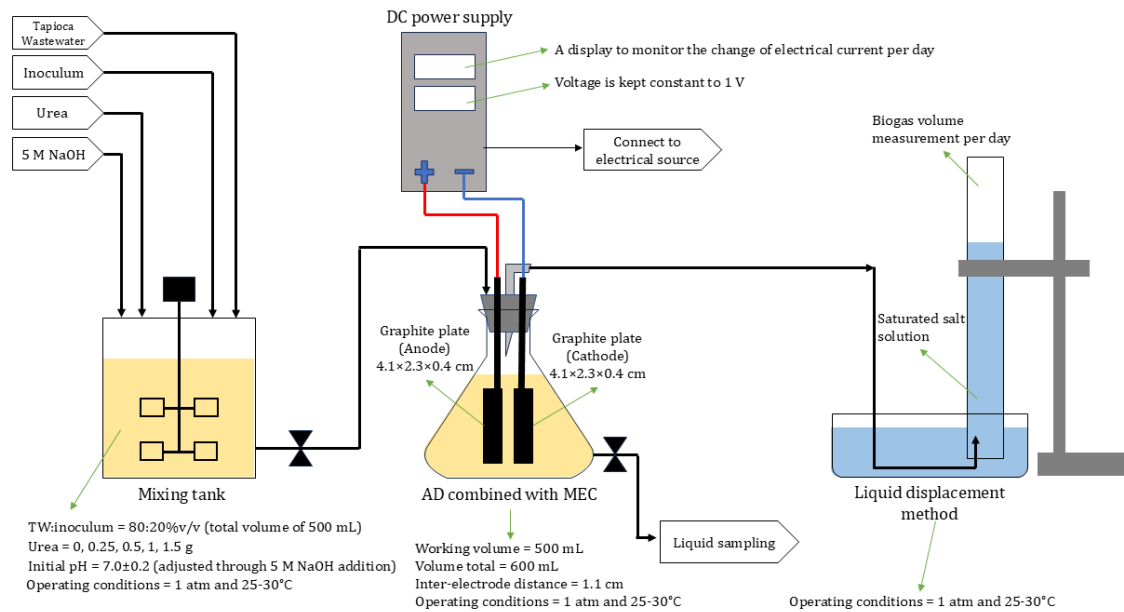


Figure 1. Laboratory scale experimental set-up.

and VFAs 385.5 mg-acetic acid/L. Fresh cow rumen fluid was sourced from a slaughterhouse located in Cilegon, Indonesia. The cow rumen fluid contains COD 26,843 mg-O₂/L and VFAs 5975.3 mg-acetic acid/L. The urea as a nitrogen source, produced by PT Pupuk Indonesia (Persero), was purchased from a local market. Black carbon graphite plates with a purity of 99.9% utilized as electrodes with a dimension of length×width×thickness of 4.1×2.3×0.4 cm were purchased from a local market.

Experimental Set-Up

Anaerobic reactors were prepared using Erlenmeyers with a total volume of 600 mL and a working volume of 500 mL. The graphite plates were located in the anaerobic digester. The positive and negative poles of the direct current power source were linked to the anode and cathode. The electrical voltage was kept constant at 1 volt. The

digester was then closed with a rubber stopper to ensure the anaerobic conditions. The liquid displacement method was used to measure the biogas that was produced. Using this method, a gas collector constructed from a reversed cylindrical glass was linked to each digester. A flexible connecting hose was used to make the connection. A detailed experimental set-up on a laboratory scale is presented in Figure 1.

Experimental Design and Procedures

A volume ratio of 80:20 v/v was used to combine rumen fluid with tapioca wastewater. Furthermore, urea addition was varied to 0.25, 0.5, 1, and 1.5 g. Then, a 5 M NaOH solution was added to adjust the liquid pH to 7.0±0.2. As control digesters, the AD alone and MEC-AD without urea were also carried out. The experimental design in this study is presented in Table 1.

Table 1. Experiment design

Digester Code	Process	Volume of Tapioca Wastewater (mL)	Volume of Rumen Fluid (mL)	Initial pH	Urea addition (g)
A	MEC-AD	400	100	7.0±0.2	0.25
B	MEC-AD	400	100	7.0±0.2	0.5
C	MEC-AD	400	100	7.0±0.2	1
D	MEC-AD	400	100	7.0±0.2	1.5
E	MEC-AD	400	100	7.0±0.2	-
F	AD	400	100	7.0±0.2	-

The anaerobic digestion process was allowed to continue at room temperature until biogas production was stopped. The biogas volume was recorded every day using the liquid displacement method. Liquid samples were collected every four days for pH, VFAs, and COD analyses. A digital pH meter was used to measure the pH of the liquid. The steam distillation method was used to assess the VFA concentration (Iqbal Syaichurrozi et al., 2024). The COD concentration was measured using the closed reflux and spectrophotometry method using a COD Reactor (Hanna H1839800) and COD meter (Hanna H183399) with High Range Plus COD reagent specifications.

RESULTS AND DISCUSSION

Biogas Production

Figure 2 presents the daily and cumulative biogas yield profiles observed throughout the research. The daily biogas production profile shown in Figure 2(a) shows the fluctuation of daily biogas yield across all variations. The optimal period for biogas generation occurred between days 2 and 12, with the highest daily biogas yield recorded in digester D at 86.6 mL/g-COD on day 3. However, a decline in daily biogas yield was observed from days 15 to 20 across all variations.

Figure 2(b) shows cumulative biogas yield. The AD and MEC-AD without urea addition

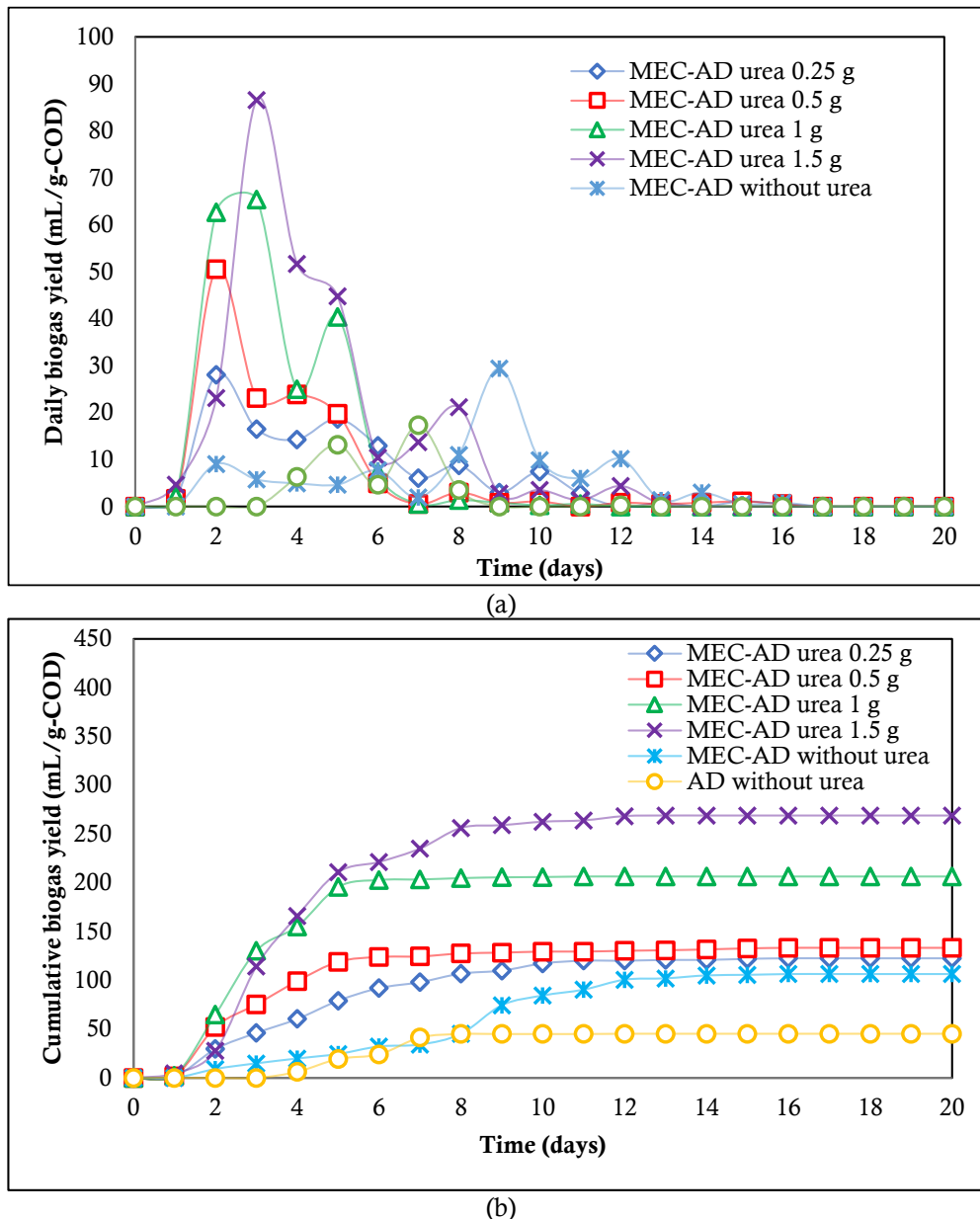


Figure 2. Biogas production profiles (a) daily, (b) cumulative.

generated a total biogas yield of 45.4 and 106.4 mL/g-COD, respectively. It proved that the presence of MEC in AD enhanced the total biogas yield by 2.3-fold compared to AD alone. The existence of MEC in AD effectively improved the rate of substrate degradation by altering the bacterial community involved in the fermentation process, thereby accelerating the digestion process and increasing biogas production (Yu et al., 2018). Some previous studies also found the same findings. Joicy et al. (2022) found that the MEC-AD in treating activated sludge waste at various voltages (0.3-0.9 V) produced 27.2%-44.8% more methane than the AD alone. Lee et al. (2017) reported that the MEC-AD of food waste generated a higher methane yield (0.34 L/g-COD) than the AD alone (0.2 L/g-COD).

The addition of the MEC to AD altered the microbial community structure and promoted electron transfer, thereby enhancing biogas production and accelerating the digestion process (Kanellos et al., 2024). In the MEC-AD process, when an electrical current was applied to the anode, an electrochemical reaction occurred, activating exoelectrogens and increasing the degradation rate of organic compounds to produce H^+ , electrons, and CO_2 (Yu et al., 2018). Exoelectrogens are a group of microorganisms that possess the ability to transfer electrons extracellularly (Logan et al., 2019). According to Wang et al. (2022), the MEC-AD process increased the number of electrogenic bacteria (exoelectrogens) by more than twofold. Exoelectrogens converted VFAs into H^+ , CO_2 , and e^- at the anode. Then, H^+ , CO_2 , and e^- were converted into CH_4 through methanogenesis based on direct interspecies electron transfer (DIET) by hydrogenotrophic methanogens at the cathode. In methane production, DIET-based methanogenesis was more efficient than methanogenesis based on indirect interspecies electron transfer (IIET) (Zhu et al., 2020). Syaichurrozi et al. (2024) stated that MEC-AD produced methane through DIET-based methanogenesis and IIET-based methanogenesis, but AD resulted in methane through IIET methanogenesis only. Hence, the existence of MEC in AD increased the conversion rate of organic compounds to methane.

The COD/N ratio is an important factor that influences the development of bacteria in breaking down organic materials during the digestion process to increase biogas production (Adiani et al., 2019). To achieve high biogas

production from tapioca wastewater, the addition of nitrogen-containing materials such as urea was necessary. At the end of the process, MEC-AD with urea addition of 0, 0.25, 0.5, 1, and 1.5 g resulted in total biogas yield of 106.4, 122.6, 133.3, 206.5, and 268.9 mL/g-COD, respectively. It showed that the highest total biogas yield was achieved with the urea addition of 1.5 g, namely 269 mL/g-COD. An increase in urea addition in MEC-AD from 0 to 1.5 g improved biogas yield from 106.4 to 268.8 mL/g-COD. Thus, the MEC-AD with urea addition of 1.5 g generated a biogas yield 2.5-fold higher than MEC-AD without urea addition. The data demonstrated that biogas production was significantly higher when urea was added compared to the control (without urea addition). This was because urea functions as a nitrogen element enhancer which was a source of nutrients for the growth of microorganisms. Urea was chosen as a nitrogen source because it was easily digested by various microorganisms (Budiyono et al., 2013). Nitrogen sources were utilized by bacteria to build their cell structures (Speece, 1983). The difference in the COD:N ratio influenced the abundance of methanogenic bacteria (Sun et al., 2018). Hence, the addition of urea in tapioca wastewater might increase the abundance of methanogenic bacteria increasing the conversion rate of COD to biogas during the MEC-AD process. In line with the current study, Budiyono et al. (2013) reported that urea addition can increase the total biogas by 52.47% compared to without urea addition.

PH and VFAs

The pH was a critical factor in anaerobic decomposition, as it significantly affected bacterial growth. The pH level in the reactor was heavily influenced by the concentrations of VFAs and ammonia/ammonium that were produced. For optimal microbial activity, the pH should be maintained between 6.5 and 8.2 (Speece, 1983). Figure 3 presents the pH profiles for each variation over 20 days.

At the beginning of the process, the initial pH of all digesters was 7.0 ± 0.2 . Based on Figure 3, the substrate pH value of digester A (MEC-AD urea 0.25 g), B (MEC-AD urea 0.5 g), C (MEC-AD urea 1 g), D (MEC-AD urea 1.5 g), E (MEC-AD without urea), and F (AD without urea) changed from pH 7 to pH 5.23, 5.62, 6.13, 6.7, 4.84, and 4.83, respectively. The decrease in substrate pH occurred from day 1 to day 8. The decrease in substrate pH

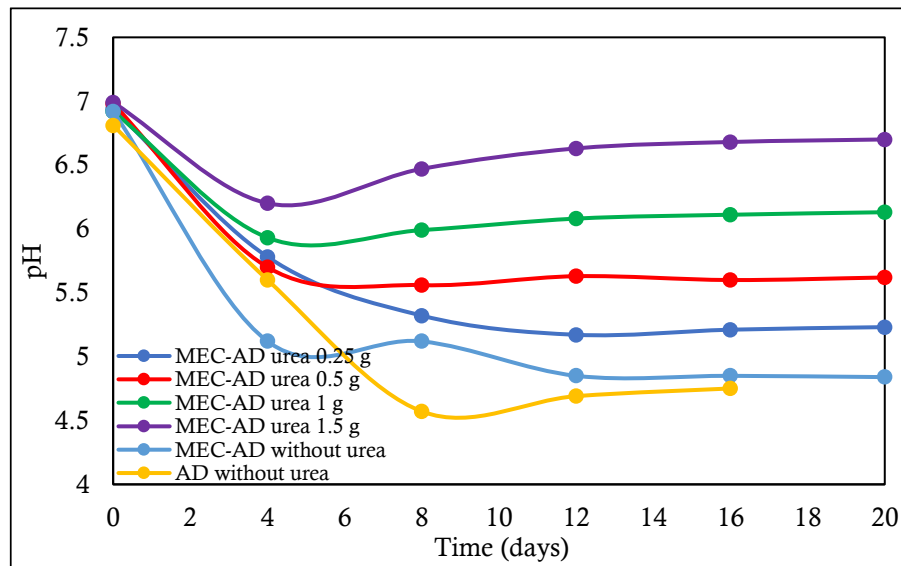


Figure 3. pH profiles.

was caused by the accumulation of VFAs which were generated during the anaerobic digestion process. This condition showed that there was an acidogenesis process in the digesters.

According to Forster-Carneiro et al. (2008), the acidogenesis occurs due to changes in hydrolysis products into VFAs. The accumulation of VFAs will reduce the pH value in the digester. Conversely, the decomposition of nitrogen into ammonia (NH_3) or ammonium (NH_4) will increase the pH value (Syaichurrozi et al., 2016). At the beginning of the process, carbohydrates are easier to be broken down than proteins so the substrate pH decreases at the beginning of the process and then increases gradually.

In digester F (AD without urea addition), there was a sharp decrease in pH at the beginning of the process (from about 7.0 to 4.57 on day 8) due to a high VFA accumulation. Such low pH levels were known to inactivate methanogenic bacteria (Sun et al., 2020). Hence, from day 9 to day 16, biogas was not formed. Methanogenic bacteria thrived in the pH range between 6.5 and 8.2 (Buyukkamaci & Filibeli, 2004).

For methanogenic bacteria to grow and develop properly, it is necessary to add nutrients to the digester. In this study, urea was added to digesters A, B, C, and D. In these digesters, the substrate pH decreased but not sharply. Urea in the system was converted to ammonia or ammonium which can maintain the substrate pH. Liu et al. (2019) revealed that protein and urea in the substrate during AD will break down into ammonia or ammonium. Ammonia when reacted with water

formed a base to provide an alkaline condition in the substrates.

The pH of the substrate was also very closely related to the amount of VFAs. Figure 4 shows the increase in VFA concentration during the 20-day fermentation period. Initially, the VFA concentration was 1503 mg-acetic acid/L in all digesters on day 0. However, after four days of fermentation, the concentrations increased to 5783 mg-acetic acid/L in digesters A and B, 3855 mg-acetic acid/L in digesters C and D, 5972 mg-acetic acid/L in digester E, and 3469 mg-acetic acid/L in digester F. This upward trend continued until day 20. Figure 4 shows that VFA concentrations in digesters A, B, C, D, and E were higher than that in digester F. This increase in VFA concentration was attributed to the presence of the MEC in AD, which enhanced the rate of substrate degradation during fermentation. According to Hassanein et al. (2017), the incorporation of MEC-AD in food waste processing can boost biogas, methane, and hydrogen production. Another research by Huang et al. (2020) found that MEC-AD significantly accelerated the degradation of carbohydrates, proteins, and VFAs by 4, 2.3, and 4.7 times, respectively, compared to AD alone. Higher VFA concentrations are crucial for increasing biogas production (Moen, 2003) because the primary component of VFAs is acetic acid, a key substrate for methanogenic bacteria in methane gas formation.

The rise in VFA concentration was also attributed to the addition of urea, which serves as a nutrient source. The presence of nitrogen sources in

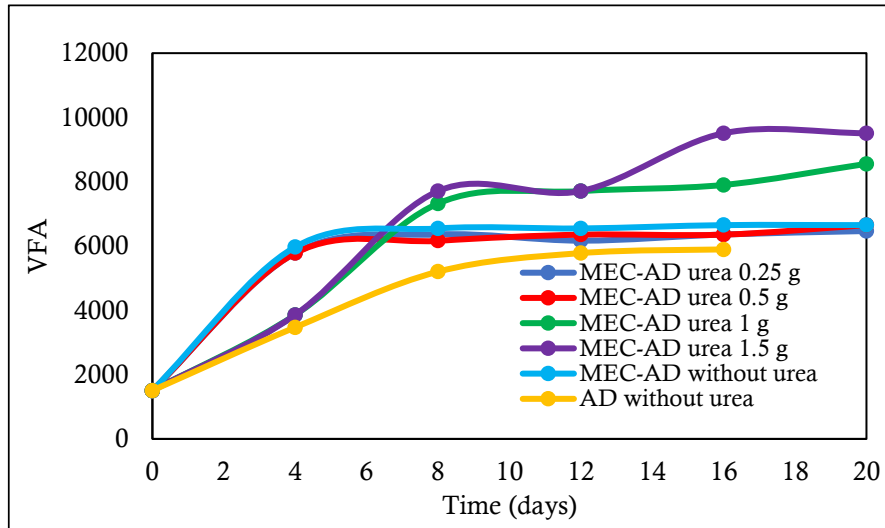


Figure 4. VFA profiles.

the substrate plays a crucial role in enhancing biogas production. Moreover, urea helps maintain a stable pH during the fermentation process, contributing to increased biogas yield. In digester D (MEC-AD urea 1.5 g), the highest VFA concentration was recorded at 9511 mg-acetic acid/L, resulting in the largest cumulative biogas volume of 269 mL/g-COD.

COD REMOVAL

The organic matter in the wastewater can be expressed as COD. The amount of COD degraded by bacteria after the fermentation process is called COD removal (Speece, 1983). The bacterial activity converts COD in the substrate into biogas, so COD can be used to calculate the potential of biogas (Rahayu et al., 2015). From the

measurement of COD levels in tapioca wastewater, it was found to be 6,419 mg-O₂/L. Furthermore, the COD level of the mixture of tapioca wastewater and inoculum was 10,504 mg-O₂/L. The COD mixture level was used as the influent COD level. The COD removal values obtained in all digesters can be seen in Figure 5.

Figure 5 shows that the COD removal in the MEC-AD digesters had a greater COD removal efficiency than that in the AD digester. This was because the existence of MEC in AD enhanced biogas production and reduced COD levels more effectively (Lee et al., 2017). From Figure 5, the digester order based on COD removal from the largest to the smallest was digester D (38%), C (35%), B (28%), A (25%), E (21%), and F (14%). The digester D had the largest COD removal

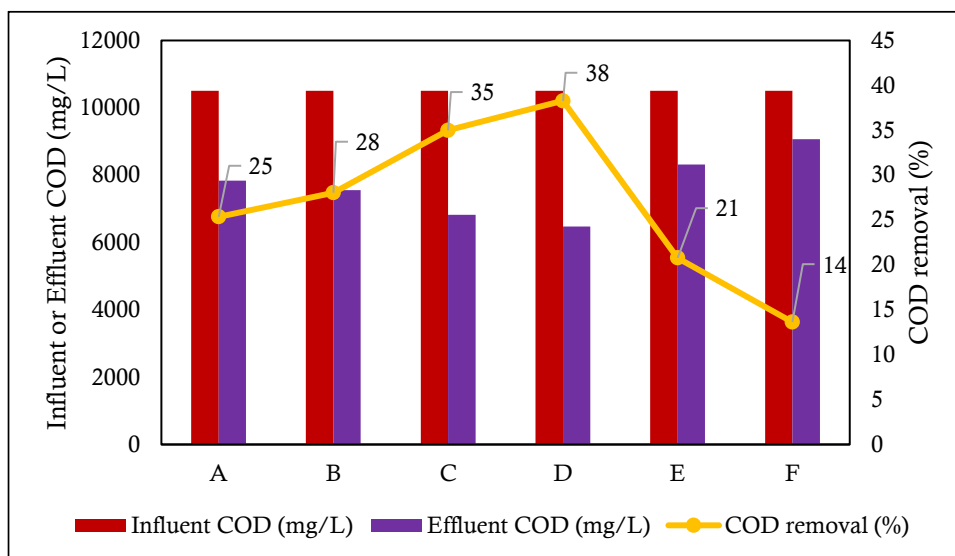


Figure 5. COD removals.

efficiency (38%) and resulted in the largest total biogas yield (268.9 mL/g-COD). The digester order based on the total biogas yield from the largest to the smallest was digesters D, C, B, A, E, and F. From these results, it can be seen that the greater the COD can be removed, the more biogas was obtained. The results of this investigation were consistent with the conclusions of a prior investigation (Widarti et al., 2015).

Limitations and Future Research

This study demonstrated that MEC-AD was a promising technology for converting tapioca wastewater into biogas. The inclusion of MEC in the AD process had a positive impact on biogas yield. In this research, the highest biogas production, 268.9 mL/g-COD, with a COD removal of 21%, was achieved in MEC-AD with the addition of 1.5 g of urea. However, methane production from MEC-AD for tapioca wastewater was still conducted at a laboratory scale using synthetic tapioca wastewater. Additionally, the composition of VFAs including acetic acid, propionic acid, butyric acid, and others, needs further verification. The detailed composition of biogas (CH_4 , H_2 , CO_2 , etc.) also requires attention and confirmation. For greater accuracy, biogas composition can be detected using gas chromatography. In future research, the MEC-AD technology to convert natural tapioca wastewater into biogas needs to be carried out on a pilot plant scale. Then, the economic analysis should be conducted in the future.

CONCLUSION

There were six digesters, namely A (MEC-AD urea 0.25 g), B (MEC-AD urea 0.5 g), C (MEC-AD urea 1 g), D (MEC-AD urea 1.5 g), E (MEC-AD without urea), F (AD without urea). Digesters A, B, C, D, E, and F generated total biogas yields of 106.4, 122.6, 133.3, 206.5, 268.9, 106.4, and 45.4 mL/g-COD, respectively. The digester order based on the total biogas yield from the largest to the smallest was digesters D, C, B, A, E, and F. It shows that MEC-AD (without urea) generated biogas yield 2.3-fold higher than AD alone (without urea). Then, in MEC-AD, the urea addition of 1.5 g generated a biogas yield 2.5-fold higher than without urea addition. The MEC-AD with urea addition of 1.5 g had the most stable substrate pH

and the highest VFAs during the process. The MEC-AD (without urea) generated a higher COD removal (21%) compared to the AD alone (without urea), namely 14%. Then, an increase in urea addition from 0 to 1.5 g in MEC-AD increased COD removal efficiency from 21% to 38%. The digester order based on COD removal from the largest to the smallest was digester D (38%), C (35%), B (28%), A (25%), E (21%) and F (14%). Hence, the greater the COD can be removed, the more biogas was obtained. In conclusion, the best variable was MEC-AD with a urea addition of 1.5 g.

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