Two step anaerobic co-digestion of sesame oil cake and sewage sludge for sludge reduction and biogas production

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Research Article

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Abstract

The aim of this study was to investigate sewage sludge reduction and biogas production using two-stage anaerobic co-digestion of sesame oil cake and sewage sludge. In the first stage (acidogenic fermentation), sesame oil cake (SOC) was acid fermented to produce fermented sesame oil cake (FSOC). In the second step (anaerobic co-digestion), sewage sludge and FSOC were mixed in various ratios of (100:0 (R1), 70:30 (R2), 50:50 (R3), and 30:70 (R4)) and observed for 30 days at a mesophilization temperature of 25 ± 2 °C. In the anaerobic co-digestion using FSOC as a co-feedstock, the VS and TS removal were in the range of 53.7 - 64.9% and 42.6 - 53.2% for R2 and R3, respectively. The highest cumulative biogas production (1740.43 mL) and methane production (1253.11 mL) were achieved with the R3. In addition, R3 had the shortest reaction delay time (λ), and stabilization of the process was the fastest of all samples. The co-digestion performance index (CPI) was determined to be 1.29, 1.39, and 1.10 for R2, R3, and R4, respectively. The highest value for R3 confirmed the highest synergistic effect. This suggests the possibility of biogas production using sesame oil cake.

Introduction

Sesame oil, which is popular in Korea, is oil made by roasting sesame seeds, and sesame oil cake (SOC) is the residue left after squeezing oil from sesame seeds. Korea's SOC production is about 3 million tons annually, the second largest quantity after rapeseed cake (ca. 6 million tons/year) (Lee et al. 2021). A portion of SOC is used as fertilizer or as a feed supplement for poultry, but most of it is disposed of as waste (Nascimento et al. 2012). Accordingly, improperly processed SOC can cause environmental problems such as the generation of harmful insects and odors. SOC consists mainly of protein (about 30%), lignocellulose (44%) and minerals (8%) (Yasothai 2014). The amount of fat in SOC varies depending on the method of squeezing the oil and the equipment used, but varies from about 5 to 11% (Nascimento et al. 2012; Yasothai 2014). The yield and composition of biogas are greatly affected by carbohydrates, fats and proteins contents of the co-feedstocks (Chanda et al. 2012). In addition, SOC contains large amounts of components such as protein, carbon, nitrogen, phosphoric acid and potassium, and it is known to have a high carbon content (C/N ratio) (Bigoniy et al. 2012; Yasothai 2014). Therefore, SOC can be an excellent candidate as a co-feedstock for biogas in anaerobic co-digestion.

In recent years, research on the synergistic effects of increasing biogas production and reducing sludge through co-digestion with various co-feedstocks and sewage sludge is underway. Co-digestion is anaerobic digestion in which various co-feedstocks and sewage sludge are combined in a single anaerobic digester (Choi 2021). Co-digestion is a viable option to increase methane yield and promote sludge reduction over anaerobic digestion with a single feedstock (Choi 2021; Jeong et al. 2019). The utilization of a co-feedstock with different chemical compositions, particularly with regard to nutrients, can improve the process due to the positive synergistic effect established in the digestion medium and the supply of scarce nutrients (Jhr et al. 2020). Co-feedstocks used for anaerobic co-digestion should be substances that can increase methanogenesis and reduce sewage sludge, and substances that can interfere with methanogenesis should be avoided (Chen and Brewer 2021; Choi 2020a). According to a recent study, a lot of waste is mixed with food waste, fish by-products and manure as a co-feedstocks for anaerobic co-digestion (Choi 2020a,b; Choi 2021; Hassan et al. 2017; Karki et al. 2021). In addition, a synergistic effect on sludge reduction and biogas production efficiency has been reported by adding biomass-based biological auxiliary raw materials such as corn stalk, coffee grounds, wheat straw, sugarcane and corn stover (Chen and Brewer 2021; Karki et al. 2021; Pohl et al. 2019; Wang et al. 2018).

Co-digestion using co-feedstocks can follow single and/or two-stage digestion systems. According to previous studies, if the single-stage co-digestion system is selected, the rapid decrease in pH due to local acid fermentation and scum conversion due to the difference in acid fermentation rate decrease the digestion efficiency of the digester (Pohl et al. 2019). In particular, ligno-cellulose, protein and fat contained in SOC can inhibit the formation of methane gas due to slow hydrolysis (Choi 2020a; Choi 2021). However, if a two-stage digestion process (acidogenic fermentation and anaerobic co-digestion) is used, the problems appearing in the single-stage co-digestion process can be addressed. In other words, in the first step (acidogenic fermentation), high molecular substances such as fat, lignin, and hemicellulose are hydrolyzed to break down into short-chain fatty acids, and monosaccharides that are easily decomposed (Choi 2020b; Pohl et al. 2019). If it is mixed with sewage sludge for anaerobic co-digestion in the second stage, it can have a synergistic effect rather than inhibiting methane gas formation and sludge reduction (Choi 2020a; Karki et al. 2021). However, to date, no research results have been found that show the production of biogas using the two-step anaerobic co-digestion process by acidogenic fermentation of sesame oil cake (FSOC) to my knowledge. Therefore, this study was conducted to observe the effect of anaerobic co-digestion in the first stage (acidogenic fermentation of SOC) and second stage (co-digestion of sewage sludge and FSOC) on biogas production and sewage sludge reduction efficiency. In addition, the synergistic effect of FSOC on anaerobic co-digestion was also observed to confirm whether it is possible to continuously use FSOC as a co-feedstock for anaerobic co-digestion.

Materials And Methods

Sewage sludge and sesame oil cake as co-feedstock

Sewage sludge was collected from the bottom of an anaerobic digester in a sewage treatment plant operated in Gangneung, Korea. The wastewater treatment plant in Gangneung is operated with a capacity of 75,000 (m^3/day) to treat domestic wastewater. The BCR (Biological Coating Removal) process is applied for sewage treatment. The daily average sludge inflow into the digester was 4,800 – 18,023 (mg/L) for TS, 6,415 – 12,657 (mg/L) for VS, and 43.4–64.7% for VS/TS. SOC was purchased as a co-feedstock from an oil mill in Gangneung city, Korea. After squeezing out the oil, the remaining hot SOC was left at room temperature for 3 hours to cool, filtered through an 80-mesh sieve to remove contaminants, and was stored in a desiccator for use in the experiment.

Experimental design

The experiment was carried out in a two-step anaerobic co-digestion process in a batch-test. The detailed experimental design of the second stage is summarized in Table 1. The first step was the acidogenic fermentation process of SOC. In the first step, after uniform mixing of SOC and tap water in a ratio of 1:3 with glucose (0.1 g/L), the mixture (40 L) was put into a 50 L cylindrical reactor, and it was stirred at 120 rpm for mesophilic digestion (35 ± 2°C) until the amount of volatile organic acids (VFAs) became constant. The prepared fermented sesame oil cake (FSOC) was stored at room temperature for use in the experiment. In the second step (anaerobic co-digestion process), sewage sludge and fermented sesame oil cake (FSOC) were mixed in various ratios of 7:3 (R2), 5:5 (R3) and 3:7 (R4). Then, the mixture (40 L) was put into a 50 L cylindrical reactor and biogas production and sewage sludge were observed for 30 days. Among the various samples, R1 consisted of only sewage sludge for use as a control. The pH was not artificially adjusted, and the temperature for mesophilization around 25°C ± 2.

Table 1
Experimental design for batch test

Samples	R1	R2	R3	R4
Injected condition	Sludge 100%	Sludge 70% + FSOC 30%	Sludge 50% + FSOC 50%	Sludge 30% + FSOC 70%
Organic loading rate (g VS_{in}/L·d)	1.43	2.26	3.82	4.65
Experimental duration	30 day			
Temperature	25°C ± 2			
Stirring speed	150 rpm			

Table 1.

Analytical methods

Total solid (TS), volatile solid (VS), and total chemical oxygen demand (TCOD) were measured based on standard test methods for water pollution processes (APHA 2012). The each test was conducted in triplicate, and the results were expressed as mean ± standard deviation. Carbon, hydrogen and nitrogen contents of Sewage sludge, SOC and FSOC samples were measured using a fully automatic 'Vario EL' element analyzer (PerkinElmer, USA). The pH was measured using a pH meter (HM-30R, DDK-TOA). For VAFs, 1 mL of the sample was centrifuged at 2000 rpm for 15 min, and the supernatant was collected and analyzed using a liquid chromatography system (1260 LC system, Agilent, USA). HPX-87H (300X 7.8 mm, Biorad, USA) was used as the analysis column. The mobile phase was 5 mM sulfuric acid (H_2SO_4), the column temperature was 60°C, and the flow rate was 0.6 (mL/min). Biogas was quantified by inserting a syringe into the rubber stopper at the top of the serum bottle at a set time every day. Biogas components (CH_4, CO_2, H_2S, H_2, NH_3, etc.) were analyzed using gas chromatography (GC, Nexis SCD-2030, Shimazu, Japan) according to ASTM D7833-14.

Mathematical modeling

Assuming that each organic material is completely decomposed into methane and carbon dioxide in the anaerobic co-digestion process, the theoretical methane yield (Y_m) was calculated by the following stoichiometric formula based on the elemental composition of organic matter.

$$\mathrm{C_aH_bO_cN_d} + \frac{(4a-b-2c+3d)}{4}\mathrm{H_2O} \rightarrow \frac{(4a+b-2c-3d)}{8}\mathrm{CH_4} + \frac{(4a-b+2c+3d)}{8}\mathrm{CO_2} + \mathrm{dNH_3}$$

1

$$Y_m = \frac{1000 \times 22.4 \times \left(\frac{4a + b - 2c - 3d}{8}\right)}{12a + b + 16c + 14d} \frac{mLCH_4}{gVS}$$

2

For the kinetic analysis of microorganisms for biogas production in the anaerobic combined process, the modified Gompertz equation was used.

$$M = M_{max}\exp\{-\exp[\frac{R_m e}{M_{max}}(\lambda - t) + 1]\}$$ (3)

Here, M is cumulative biogas production (mL/g·VS), M_{max} is biogas yield potential (mL/g·VS), R_m is the maximum biogas production rate (mL/g·VS·d), λ is the duration of the lag phase (days), t is the time (d) at which cumulative methane production M is calculated (days) and *e* is Euler's number. CPI (co-digestion performance index) was calculated using the following formula.

$$CPI = \frac{B_P}{B_{VS}}$$

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Here, B_p is the bio-methane potential of the co-digestion blend, B_{vs} (= $\sum_i^n \%VS_i B_{ip}$) is the weighted average based upon VS content (%VS) of the individual substrate bio-methane potentials (B_{ip}). Substrate *i* through *n* are co-digested such that $\sum_i^n \%VS_i = 1$. Thus, a CPI > 1 indicates a synergistic effect of co-digestion, and CPI < 1 indicates an antagonistic effect.

To select appropriate kinetic model for anaerobic co-digestion processes, Chi-squared (χ^2) was calculated using Eq. (5).

$$\chi^2 = \sum_{i=1}^{n} \frac{(M_{exp} - M_{cal})2}{M_{cal}}$$ (5)

where M_{exp} and M_{cal} are methane production obtained from experiments and models, respectively.

Results And Discussion

Properties of samples

Properties of co-feedstock

The comparative analysis of the components of SOC and FSOC was summarized in Table 2. In anaerobic co-digestion, the carbon and nitrogen contents (C/N) affecting the growth of microorganisms increased in FSOC more than in SOC. The C/N ratio of SOC increased after fermentation stage from 6.43 to 16.87. The content of VS also increased and the change of favorable components from SOC to FSOC, which affects biogas production in anaerobic co-digestion.

Table 2
Properties of co-feedstocks

	C (%)	H (%)	N (%)	C/N	VS (%)	TS (%)
Sewage Sludge	37.13	5.72	6.61	5.64	15.72	21.33
SOC	43.24	7.12	6.72	6.43	63.22	93.46
FSOC	52.63	6.32	3.12	16.87	86.41	90.33

Previous study reported that biogas production increased by 3–5 times when VS consumed by 2 times (Chanda et al. 2012; Choi 2020b). This is an important reason for performing two-step anaerobic digestion without directly incorporating SOC into the anaerobic tank.

Table 2.

Change of pH and VFAs

The change in VFA according to pH was in Fig. 1. The fermentation broth of SOC reached pH 4 after 5.5 days of reaction, and the amount of VFAs at pH 4 remained almost unchanged. Therefore, it was judged that the reaction for the production of FSOC in which SOC was fermented on day 6 was completed. The average pH of SOC was 6.8, whereas the pH decreased with increasing VFAs in FSOC. The prepared FSOC was mixed with the sewage sludge of the anaerobic digester according to the experimental design, and the initial pH showed various values (6.8–7.5) depending on the mixing ratio of the sewage sludge and the FSOC.

Figure 1.

It has been reported that most anaerobic microorganisms, including methanogens, have the best activation within the pH range of 6.8 to 8.5 (Chen and Brewer 2021; Choi 2020b). The pH of the anaerobic digester initially decreased due to the production of volatile acids. However, the pH of the digester increased and stabilized as the methanogen consumed volatile acids and alkalinity was created (Chanda et al. 2012; Choi 2020a). With a residence time of about 5 days or more, the methane-forming methanogens begin to consume the volatile acids rapidly. In a properly functioning anaerobic digester, the pH is maintained between 6.8 and 7.2 as the volatile acids are converted to methane and carbon dioxide. The pH of anaerobic systems is strongly influenced by the carbon dioxide content of the biogas. Therefore, a two-step co-digestion process is advantageous to shorten the biogas production lag time and promote the initial activation of methanogens.

Characterization of samples

Because the growth rate of anaerobic microorganisms for biogasification is slow, the rate is greatly affected by changes in the operating parameters of the anaerobic digester. Therefore, for stable operation of anaerobic co-digestion, it is important to maintain/manage operating factors such as pH, C/N ratio, temperature, and VFAs/alkalinity within an appropriate range (Chen and Brewer 2021; Wang et al. 2018). In addition, when ammonia and heavy metals exceed a certain concentration, close attention is required because the proliferation and activity of anaerobic microorganisms is inhibited by the dissociation equilibrium and toxicity in the anaerobic digester (Choi 2021; Choi 2020a). The microorganisms in the anaerobic digester show different activities depending on the pH range, and since the dissociation equilibrium of inhibitors such as ammonia and

VFAs is controlled by the measured pH, it is necessary to maintain an appropriate pH in consideration of the pH value of the organic waste. The pH of the samples ranged from 6.8 to 7.5, and the pH was the lowest in R4, which had the highest content of FSOC (Table 3). The pH of the anaerobic digester initially decreased due to the production of volatile acids, but alkalinity was created as the methanogenic bacteria consumed the volatile acids. Then, the pH of the digester increased, and the process stabilized.

Table 3
Characteristics of digesters with different co-substrates ratios

	R1	**R2**	**R3**	**R4**
pH	7.5 ± 0.4	7.3 ± 0.3	7.0 ± 0.2	6.8 ± 0.2
Total Solids (TS) (%)	21.3 ± 0.1	42.7 ± 2.1	64.1 ± 1.8	85.5 ± 2.3
Volatile Solids (VS) (%)	15.7 ± 0.4	36.3 ± 1.9	56.9 ± 2.2	77.5 ± 1.2
VS/TS	73.71 ± 0.02	85.01 ± 0.02	88.77 ± 0.02	90.64 ± 0.02
Total carbon (%, dry weight)	26.8 ± 0.5	46.6 ± 0.9	52.2 ± 1.5	58.2 ± 1.2
Total nitrogen (%, dry weight)	4.2 ± 0.1	4.0 ± 0.3	3.9 ± 0.2	3.8 ± 0.4
C/N ratio	5.64 ± 0.1	11.7 ± 0.3	13.4 ± 0.4	15.3 ± 0.2
Alikalinity (g as $CaCO_3$/L)	2.48 ± 0.09	1.99 ± 0.05	1.69 ± 0.05	1.46 ± 0.05
Volatile Fatty Acids (VFAs) (g/L)	0.46	0.76	1.12	1.89
VFAs/Alkanity	0.19	0.38	0.66	1.29

The C/N ratio of the samples was in the range of 5.6–15.3, and the C/N ratio increased as the FSOC content increased. The C/N ratio for the growth of microorganisms is an important factor influencing anaerobic digestion. According to previous studies, the appropriate C/N ratio for anaerobic digestion using sewage sludge is in the range of 15–30 (Choi 2020a; Hassan et al. 2017). However, when using a co-feedstock, the appropriate range of C/N ratio may vary depending on the type of co-feedstock. For example, in the anaerobic co-digestion process, cattle manure, poultry manure, pig manure and sheep manure have a relatively low C/N ratios of 7–34. In contrast, rice straw, wheat straw, corn waste, and sugarcane have high C/N ratios of 51–151 (Chanda et al. 2012; Siddique and Wahid 2018). Thus, the C/N ratio varies according to the type of co-substrate in anaerobic co-digestion. In this study, VFAs were measured as 0.46–1.89 (g/L), which is not considered to be a limiting factor in methane gas formation. The VFAs/alkalinity was calculated in the range of 0.19–1.29, which corresponds to an appropriate ratio except for the R4 sample. VFAs are organic acids generated by the decomposition of organic compounds, but the buffering capacity of the fermentation substrate decreases when they excessively accumulate inside the anaerobic digestion tank (Cheng and Brewer 2021; Strazzera et al. 2018). The concentration of VFAs that inhibits methane formation differs depending on the co-feedstocks. According to a previous study, when the concentration of VFAs is above 4000 mg/L and the ratios of VFAs and alkalinity are above 0.8 in anaerobic co-digestion, the pH decreases and methane production is inhibited

(Chanda et al. 2012; Choi 2021). However, the ratio of VFAs/alkalinity was determined above 1.2 using food waste in an anaerobic co-digestion process (Jeong et al. 2019; Karki et al. 2021), but the pH and biogas production did not decrease. Therefore, it is necessary to optimize the ratio of VFAs and alkalinity according to the type of auxiliary substrate and the reaction conditions.

Table 3.

Sludge reduction

Figure 2 shows the total solids (TS) and volatile solids (VS) removal efficiencies in the anaerobic co-digestion process using sewage sludge and FSOC. The TS removal efficiencies of R1, R2, R3 and R4 averaged 28.5%, 42.6%, 53.2% and 34.8%, respectively, while the VS removal efficiencies were 37.6%, 53.7%, 64.9% and 44.9%, respectively. The TS removal efficiency was lower than that of the VS for 30 days in anaerobic co-digestion. In particular, R1, which was anaerobically digested with only sewage sludge, showed the lowest removal efficiency, and the R3 sample showed the highest removal efficiency of TS and VS. Chanda et al. (2012) reported an average VS removal efficiency of 53.6% for 30 days in an anaerobic co-digestion process using jatropha oil seed cake. This result was similar to R2 but has a lower removal efficiency than R3. This is thought to be because jatropha oil cake contains a higher concentration of long-chain fatty acids compared to FSOC. On the other hand, FSOC decomposed long-chain fatty acids into short-chain fatty acids through a two-step digestion process. In summary, this study showed that the anaerobic co-digestion of FSOC showed higher removal efficiency of TS and VS than that of using only sewage sludge. This was is a very attractive advantage of anaerobic co-digestion using FSOC.

Figure 2.

Biogas production

Methane and carbon dioxide content of produced biogas

The cumulative biogas production volumes in the anaerobic co-digestion for R1, R2, R3 and R4 were 399.21, 1380.22, 1740.43 and 892.99 mL, respectively (Fig. 3). The specific biogas production (mL/g·VS) ranges were 0.01–1.78 for R1, 0.01–3.27 for R2, 0.01–3.59 for R3 and 0.01–0.93 for R4, and the average biogas vs. VS in all samples was observed in a range of 0.59–2.04 (mL/g·VS) (Fig. 4). The production of biogas was low until 3 days after the reaction, and it started to increase after 5 days of the reaction in R1 and R4 samples. In particular, the biogas production increased significantly after 5 days for R2 and R3 samples, and the maximum cumulative biogas yield was observed for R3. The yield and composition of biogas is greatly affected by the carbohydrate, fat and protein content of the co-feedstock (Chanda et al. 2012). The critical toxicity level of inorganic substances in anaerobic co-digestion depends on whether the co-feedstocks are acting alone or in combination (Strazzera et al. 2018). That is, certain combinations may have synergistic effects, while other combinations may have antagonistic effects. It was confirmed that co-digestion using FSOC as a co-feedstock has a favorable effect on biogas production.

The average cumulative methane volumes produced over 30 days of the anaerobic co-digestion process were 207.59, 924.75, 1253.11 and 509.00 mL for R1, R2, R3 and R4, respectively, and the methane content of the produced biogas was determined to be in a range of 51.91–72.06% for all samples. The maximum average methane content reached 72.06% for R3, and the minimum average methane content was 51.91% for R1. In R2 and R3, both the process stabilization time was fast, and the biogas production was high, and the content of methane gas was also higher than that of R1 and R4 samples. The carbon dioxide content of the produced biogas was measured in the range of 127.75–348.09 mL for all samples, which was in the range of 20–32% of produced biogas. Sample R1 showed the highest ratio of 32%, and R3 was the lowest with 20%. According to the report of previous studies, the methane content was 60.7–68.0% and carbon dioxide content was 31.7–38.3% in biogas from the anaerobic co-digestion of jatropha oil cake as a co-feedstock (Chanda et al. 2012). After process stabilization, the average proportion of methane produced from biogas measured by Deshpande et al. (2012) ranged from 68 to 72%, and the amount of carbon dioxide was 28 to 31%. Similar results for the range of methane concentration of biogas in anaerobic co-digestion using various co-feedstocks were also observed in refs. (Chanda et al. 2012; Deshpande et al. 2012; Jhr et al. 2020; Pohl et al. 2019; Wang et al. 2018).

Figure 3 **and** Fig. 4.

Anaerobic co-digestion can produce synergistic interactions through nutrient balance, trace element supplementation, dilution of toxic and inhibitory compounds, and promotion of microbial diversity (Pan et al. 2019). FSOC also has a high content of nutrients and trace elements, so it increases the diversity of anaerobic microorganisms in anaerobic co-digestion, prevents partial accumulation of VFAs in the digester, and activates methanogenic bacteria at a balanced C/N ratio for biogas production (Esteban-Gutiérrez et al. 2018; Hassan et al. 2017; Pan et al. 2019). A combination of substrates through anaerobic co-digestion can lead to synergistic effects such as dilution of methanogenesis inhibitors, addition of nutrients to increase biodegradability and/or changes in the microbiome resulting in improved metabolism (Chen and Brewer 2021; Pan et al. 2019). Ebner et al. (2016) proposed comparing the biomethane potentials of co-digested substrates with a weighted sum of single-substrate biomethane potentials as a measure of synergistic or adversarial interactions. The co-digestion performance index (CPI), or synergy index, is defined as the specific methane yield (SMY) due to co-digestion divided by the weighted average of the SMY obtained from a single digestion of each co-feedstock (Pan et al. 2019). A high CPI value does not guarantee the maximum SMY if it is used as a performance indicator. Therefore, both SMY and CPI should be considered to determine the optimal mixing ratio of the common co-feedstock. CPI is used to evaluate antagonistic (CPI < 1), additive (CPI = 1) and synergistic (CPI > 1) interactions in co-digestion (Karki et al. 2021). The calculated CPI was 1.29 for R2, 1.39 for R3, and 1.10 for R4, indicating that R3 had the highest synergistic effect, and R1 was used as a control (Fig. 5).

According to a previous study, when pig manure: corn stover: cucumber residues were used as co-feedstocks, the CPIs were determined to be 1.9 and 1.8, respectively, indicating a high synergistic effect (Wang et al. 2018). On the other hand, when food waste: toilet paper and oat straw: cow manure were used as co-feedstocks, CPIs were − 1.0 and − 1.5, respectively, and no synergistic effect was found in anaerobic co-digestion (Kim et al. 2019; Zhao et al. 2018). As such, the synergistic effect for biogas production is determined depending on which co-feedstock is used for anaerobic co-digestion.

Figure 5.

Table 4 summarizes the amount of biogas produced using various co-feedstocks. Sugarcane straw, wheat straw, rice straw and fallen leaves showed low methane production of 0.067–0.174 (mL/g·VS), whereas Jatropha and sunflower oil cakes reached high methane production in the range of 0.422–0.460 (mL/g·VS). De-oiled seed cakes have a high VS content, including hard lignocellulose, proteins, digestible carbohydrates and lipids (Jhr et al. 2020), with the lipid content contributing the most to CH_4 formation. Higher operating temperatures, lower substrate concentrations and lower lignin content provide higher biogas yields. Although the oil seed cake contains some lignin, most of the lignocellulosic matrix degrades during oil extraction, resulting in a low C/H ratio (9–12) and a high CH_4 yield (Choi 2020a; Paritosh et al. 2021). In this study, 0.56 (mL/g·VS) of methane production was measured for R3, which produced about twice as much methane gas than sugarcane straw or fallen leaves.

Table 4
Biogas yield by various co-feedstock

Feedstock	Operating conditions		CH_4 Yield (mL/gVS)	Reference
	C/N	Time (day)		
Wheat straw	53.5	40	0.1779	Hassan et al. 2017
Rice straw	59.5	30	0.1743	Kaintholaet al. 2019
Fallen leaves	58.9	30	0.0673	Elsayed et al. 2021
de-oiled rice bran	16.5	40	0.270	Jhr et al. 20208
Pig manure	13.0	85	0.3587	Wang et al. 2018
Suldge and fishwaste (7:3)		30	0.38	Choi 2020a
Food waste	15.1	30	0.447	Tang et al. 2019
Jatropha oil cakes	12.7	30	0.422	Chanda et al. 2012
Pongamia oil cakes	8.7	30	0.448	Chanda et al. 2012
Sunflower oil cake	11.8	7	0.460	Raposo et al. 2009
sewage sludge and fermented sesame seed oil cake (7:3)	11.7	30	0.42	This study
sewage sludge and fermented sesame seed oil cake (5:5)	13.4	30	0.56	This study

Lignocellulosic biomass feedstock has great potential in anaerobic co-digestion due to its abundance, low cost and high availability throughout the year. However, the slow hydrolysis rate of lignocellulosic-based biomass in general limits the single digestion of these highly recalcitrant feedstocks, which is often addressed by utilizing expensive pretreatment (Chen and Brewer 2021; Paritosh et al. 2021). Pretreatment to improve the stability of the anaerobic process has been used to loosen the structure of the biomass, release more readily degradable components, increase the accessible surface area, and remove toxic saponins from the oil seed cake (Chen and Brewer 2021; Grübel et al. 2019). In particular, some methanogenesis-inhibiting compounds, such as furfural and hydroxymethyl-furfural, are inevitably released from pretreatment of lignocellulosic biomass (Jhr et al. 2020; Karli et al. 2021; Monlau et al. 2013), meaning that not all pretreatments have a positive effect on CH_4 production. It was recently reported that CH_4 production was improved by more than 17% using an efficient two-stage anaerobic co-digestion (Chen and Brewer 2021; Karki et al. 2021; Pohl et al. 2019). According to a previous study, methanogenesis inhibiting compounds were not significantly produced in the two-step anaerobic digestion (Choi 2020a; Paritosh et al. 2021; Pohl et al. 2019). In addition, two-stage anaerobic co-digestion can reduce the use of chemicals (Choi 2020a,b; Siddique and Wahid 2018).

Table 4.

OLR (Organic loading rate)

OLR refers to the amount of organic matter supplied to anaerobic co-digestion, and excessively high OLR organic or inorganic substances inhibit bacterial growth (Choi 2020a; Pohl et al. 2019). For various samples, the cumulative biogas production increased from 399.21 to 1740.43 (mL/m^3) when the OLR increased from 1.43 to 3.82 ($g{\cdot}VS_{in}/L{\cdot}d$) and the COD loading rate increased from 5.67 to 14.72 ($gCOD_{in}/L{\cdot}d$) (Table 5). The COD loading rate was higher than the VS loading rate in all samples. However, cumulative biogas production decreased in R4, which showed the highest organic content among samples with an OLR of 4.65 ($gVS_{in}/L{\cdot}d$) and a COD loading rate of 16.18 ($gCOD_{in}/L{\cdot}d$). Since OLR is a major factor influencing the removal efficiency of VS or the productivity of biogas, many researchers have conducted various studies to find the optimal OLR (Cheng and Brewer 2021; Choi 2020a). According to previous studies, the optimal OLR of the anaerobic co-digestion is about 3–4 ($kg{\cdot}VS/m^3{\cdot}day$), and the biogas production decreases when it exceeds 4.2 ($kg{\cdot}VS/m^3{\cdot}day$) (Cheng et al. 2020; Paritosh et al. 2021). Jeong et al. (2019) reported that the maximum operable OLR in a single plug flow anaerobic digester was (4.2 $kg{\cdot}VS/m^3{\cdot}day$), and Hassan et al. (2017) suggested 3 ($kg{\cdot}VS/m^3{\cdot}day$) as the optimal OLR condition for the integrated digestion of goose manure and straw in a single CSTR reactor. The optimal OLR of anaerobic co-digestion depends on a number of parameters, including the nature of the feed organism, operating temperature and reaction time (HRT) (Choi 2021; Choi 2020a). Previous studies have shown that a balanced C/N ratio achieved through simultaneous anaerobic co-digestion of different feedstocks prevents the accumulation of VFAs due to the improved buffering capacity despite the high OLR (Choi 2020a; Hassan et al. 2017; Strazzera et al. 2018). For example, anaerobic co-digestion of food waste and pig wastewater, which are rich in trace elements or nutrients, can prevent VFA accumulation, resulting in stability and improved methane production rates (Esteban-Gutiérrez et al. 2018; Wang et al. 2018). Although food waste is deficient in trace elements, it plays an important role in activating the enzymes necessary for the growth of synthetic trophic bacterial communities and methanogens (e.g., carbon monoxide dehydrogenase, coenzyme M-methyltransferase complex, and coenzyme F430) (Latha et al. 2019). On the other hand, nutritional balance and supplementation of trace elements (e.g., Fe, Ni, Co, etc.) by pig wastewater can enhance the microbial diversity and enzymatic activity and support symbiotic and neotrophic associations (Aboudi et al. 2015; Wang et al. 2018). In general, a low OLR can cause nutritional deficiencies in the fermenting microorganisms, which can lead to low anaerobic digestion efficiency (Latha et al. 2019; Zhang et al. 2019). Conversely, a OLR that is too high can cause accumulation of VFA in the digester, and consequently inhibit the activation of bacteria, thereby causing a decrease in process efficiency (Aboudi et al.

2015; Choi 2020a). As a result of this study, it can be seen that co-digestion using FSOC is suitable for biogas production using wastes with high organic matter content because stable operation is possible even under high OLR conditions (R3 is 3.82) compared to a general single digester. In conclusion, the treated substrate in the anaerobic co-digestion using FSOC both produces a larger amount of biogas and significantly reduces the contents of TS and VS in the digester.

Table 5
Organic loading rate and biogas production by various samples

Samples	OLR ($gVS_{in}/L·d$)	COD loading rate ($gCOD_{in}/L·d$)	Biogas production ($L/gVS_{in}·d$)	Cumulative biogas production (mL)
R1	1.43	5.67	0.85	399.21
R2	2.26	11.36	1.02	1380.22
R3	3.82	14.72	1.27	1740.43
R4	4.65	16.18	0.38	892.99

Table 5.

Kinetic data

An analysis of the kinetics during anaerobic co-digestion was conducted to find the optimal conditions for biogas production by analyzing the mechanism of microbial activity and removing factors that interfere with the process (Choi 2020b). Since the production of biogas is closely related to the growth and activation of methane microorganisms, the produced biogas is a result of the growth and activation of microorganisms (Choi 2020a). The modified Gompertz equation (Eq. 3) was used for the kinetic analysis of microorganisms for biogas production in the anaerobic co-digestion process, and the results are summarized in Table 6. The λ (d) representing the reaction delay time was 5.86, 2.89, 2.33 and 3.61 for R1, R2, R3 and R4, respectively, with R1 being the longest and R3 being the shortest. In particular, R2, R3 and R4 samples using FSOC had shorter reaction delay times for biogas production compared with R1 using only sewage sludge. These results confirm that FSOC improves the biodegradability of sewage sludge, and it is also demonstrated that methanogenic bacteria are activated in a short time in anaerobic co-digestion. The values of M and R_{max} were inversely proportional to the value of λ. In other words, methanogenic microorganisms were activated earlier for shorter reaction delay times, which led to an increase in the methane production. These results are also related to the value of C/N ratio mentioned above. The ratio of the value of M to the M_{max} value, which can be interpreted as the conversion efficiency of organic matter into methane in anaerobic co-digestion, was in the range of about 97.80-99.34% when using FSOC. This was about 10% higher than the case where only sewage sludge was used (87.88%). These results showed that there is a way to achieve stable digestion conditions by balancing the nutritional status in anaerobic co-digestion of FSOC. The correlation coefficients (R^2) showed high significance for all samples in the range of 0.9907–0.9989. In particular, R3 showed the highest correlation of 0.9989. Considering these results, anaerobic co-digestion in which FSOC is acid-fermented and combined with sewage sludge is a new useful method that can simultaneously treat waste and produce biogas.

Table 6
Kinetic parameters from different models in anaerobic co-digestion

Samples	M_{max}	R_{max}	λ	M	χ^2	R^2
R1	207.59	21.62	5.86	182.24	1.9362	0.9907
R2	924.75	58.43	2.89	918.65	1.5011	0.9963
R3	1253.11	64.57	2.33	1236.42	1.2734	0.9989
R4	509.01	37.19	3.61	497.83	1.8344	0.9923

M is the cumulative methane yield (mL of CH_4/d) at time t, M_{max} is the maximum methane formed (mL of CH_4/d), R_{max} is the maximum rate of methane production (mL of CH_4/d), e is Euler's constant, λ is the lag phase constant (d)

Table 6.

Stability indicators of co-digestion and estimation of energy balance

Energy-economic calculations for anaerobic co-digestion using FSOC may affect the feasibility of anaerobic co-digestion and may influence the assessment of whether FSOC can be used continuously as a co-feedstock (Karki et al. 2021; Zhang et al. 2019). Table 7 summarizes an energy-economic analysis that was performed using methane gas produced when biogas was produced by anaerobic co-digestion using FSOC. The amount of energy recoverable was calculated by measuring the amount of methane in the total amount of biogas generated and using the calorific value of methane. The amount of methane produced per input VS showed the highest value in R3, and thus the amount of energy recovery was also highest in R3 at 5.61 (kWh), followed by R4 at 4.21 (kWh). R4 and R1 showed values of 2.51 and 3.21 (kWh), respectively, and R1 showed the lowest energy production. In particular, R3 produced about 2.24 times more energy than that of R1, so a ratio of 5:5 is recommended when biogas is produced by anaerobic co-digestion of FSOC. Combining the experimental results of biogas generation, organic matter removal, and methane content in biogas, it was found that when sewage sludge and FSOC were mixed in anaerobic co-digestion, more energy could be recovered compared to the anaerobic process using only sewage sludge.

Table 7
CH_4 and energy production according to mixing ratio

Samples	CH_4 production	Energy Production	
	($m^3CH_4/kg \cdot VS$)	($kcal/kg \cdot VS$)[a)]	($kWh/kg \cdot VS$)[b)]
R1	0.25	2155.0	2.51
R2	0.42	3620.4	4.21
R3	0.56	4827.2	5.61
R4	0.32	2758.4	3.21
[a)] CH_4 production x 8,620 kcal/m^3 CH_4, [b)]1kcal = 1.163Wh			

Table 7

Conclusions

Sewage sludge reduction and biogas production were measured in a two-step anaerobic co-digestion process with sesame oil cake. The C/N ratio increased from 6.43 for SOC to 16.87 for FSOC, and the content of carbon and hydrogen that affects the growth of microorganisms in anaerobic co-digestion increased in FSOC rather than in SOC. The average TS removal reached 28.5, 42.6, 53.2 and 34.8%, while VS removal was 37.6, 53.7, 64.9 and 44.9% for R1, R2, R3 and R4, respectively. The cumulative biogas volumes produced in R1, R2, R3 and R4 were 399.21, 1380.22, 1740.43 and 892.99 (mL), respectively. In addition, the cumulative methane production during 30 days of the anaerobic co-digestion process was confirmed to be 51.91–72.06% of the produced biogas. The CPI values were 1.29 for R2, 1.39 for R3, and 1.10 for R4, indicating that R3 had the highest synergistic effect. In terms of the reaction delay time λ (d), R1, R2, R3 and R4 showed values of 5.86, 2.89, 2.33 and 3.61, respectively. Finally, the amount of energy recovery in R3 was the highest at 5.61 (kWh). Therefore, when producing biogas by anaerobic co-digestion of FSOC, a ratio of 5:5 is recommended. Overall, the anaerobic co-digestion of FSOC can recover more energy and further reduce sludge compared to an anaerobic process using only sewage sludge based on the combined experimental results of biogas production, organic matter removal and methane content in biogas.

Declarations

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Author contribution

HJ Choi: Conceptualization, literature search, literature analyze, design, data, analysis, writing and revision of the manuscript. All authors agreed with the content and all gave explicit consent to submit the manuscript. This manuscript describes work, which has not been published before and is not under consideration by any other journal.

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Data availability

All data generated or analysed during this study are included in this article.

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References

1. Aboudi, K., Álvarez-Gallego, C.J., Romero-García, L.I.: Semi-continuous anaerobic co-digestion of sugar beet byproduct and pig manure: effect of the organic loadingrate (OLR) on process performance. Bioresour Technol. **194**, 283–290 (2015)
2. APHA(2012) Standard methods for the examination of water and wastewater, 22nd edition edited by E. W. Rice, R. B. Baird, A. D. Eaton and L. S. Clesceri. American Public Health Association (APHA), American Water Works Association (AWWA) and Water Environment Federation (WEF) Washington, D.C., USA
3. ASTM D7833–14, Standard Test Method for Determination of Hydrocarbons and Non-Hydrocarbon Gases in Gaseous Mixtures by Gas Chromatography, Active Standard ASTM D7833 | Developed by Subcommittee: D03.07, Book of Standards Volume: 05.06, Https://www.astm.org/Standards/D7833.html
4. Bigoniy, P., Nishad, R., Singh, C.S.: Preventive effect of sesame seed cake on hyperglycemia and obesity against high fructose-diet induced Type 2 diabetes in rats. Food Chem. **133**, 1355–1361 (2012)
5. Chanda, R., Vijay, V.K., Subbarao, P.M.V., Khura, T.K.: Production of methane from anaerobic digestion of jatropha and pongamia oil cakes. Appl. Energy **93**, 148–159 (2012)
6. Cheng, F., Bayat, H., Jena, U., Brewer, C.E.: Impact of feedstock composition on pyrolysis of low-cost, protein- and lignin-rich biomass: A review. J. Anal. Appl. Pyrolysis **147**, 104780 (2020)
7. Cheng, F., Brewer, C.E.: Conversion of protein-rich lignocellulosic wastes to bio-energy: Review and recommendations for hydrolysis + fermentation and anaerobic digestion. Renew. Sustain. Energy Rev. **146**, 111167 (2021)
8. Choi, H.J.: Acid-fermented fish by-products broth: An influence to sludge reduction and biogas production in an anaerobic co-digestion. J. Environ. Manag **262**, 110305 (2020a)
9. Choi, H.J.: Assessment of sludge reduction and biogas potential from anaerobic co-digestion using an acidogenically fermented fishery byproduct with various agricultural wastes. Water Air Soil Pollut **231**(336),

2–12 (2020b)

10. Choi, H.J.: Influence of acidogenic fermented fish by-products with rice bran for sludge reduction and biogas recovery in anaerobic co-digestion. Environ. Eng. Res. **26**(1), 190409 (2021)
11. Deshpande, N.V., Kale, N.W., Deshmukh, S.J.: A study on biogas generation from Mahua (Madhuca indica) and Hingan (Balanites aegyaptiaca) oil seedcake. Energy Sustain. Dev. **16**, 363–367 (2012)
12. Ebner, J.H., Labatut, R.A., Lodge, J.S., Williamson, A.A., Trabold, T.A.: Anaerobic co-digestion of commercial food waste and dairy manure: Characterizing biochemical parameters and synergistic effects. Waste Manag **52**, 286–294 (2016)
13. Elsayed, M., Blel, W., Soliman, M., Andres, Y., Hassan, R.: Semi-continuous co-digestion of sludge, fallen leaves, and grass performance. Energy **221**, 119888 (2021)
14. Esteban-Gutiérrez, M., Garcia-Aguirre, J., Irizar, I., Aymerich, E.: From sewage sludge and agri-food waste to VFA: Individual acid production potential and up-scaling. Waste Manag **77**, 203–212 (2018)
15. Grübel, K., Kuglarz, M., Waclawek, S., Padil, V.V.T., Černik, M., Varma, R.S.: Microwave-assisted sustainable co-digestion of sewage sludge and rapeseed cakes. Energy Convers. Manag **199**, 112012 (2019)
16. Hassan, M., Ding, W., Umar, M., Rasool, G.: Batch and semi-continuous anaerobic co-digestion of goose manure with alkali solubilized wheat straw: A case of carbon to nitrogen ratio and organic loading rate regression optimization. Bioresour Technol. **230**, 24–32 (2017)
17. Jeong, J., Kang, H., Kim, M.: Development of integrated system coupled with anaerobic kinetic phase separation and in-situ methane enrichment system for biomethane production. J. Korea Soc. Waste Manag **36**(3), 250–259 (2019)
18. Jhr, B., Chandra, R., Vijay, V.K., Subbarao, P.M.V., Isha, A.: Utilization of de-oiled rice bran as a feedstock for renewable biomethane production. Biomass Bioenerg **140**, 105674 (2020)
19. Karki, R., Chuenchart, W., Surendra, K.C., Shrestha, S., Raskin, L., Sung, S., Hashimoto, A., Khanal, S.K.: Anaerobic co-digestion: Current status and perspectives. Bioresour Technol. **330**, 125001 (2021)
20. Kainthola, J., Kalamdhad, A.S., Goud, V.V.: Enhanced methane production from anaerobic co-digestion of rice straw and hydrilla verticillata and its kinetic analysis. Biomass Bioenerg **125**, 8–16 (2019)
21. Kim, J., Kim, J., Lee, C.: Anaerobic co-digestion of food waste, human feces, and toilet paper: Methane potential and synergistic effect. Fuel **248**, 189–195 (2019)
22. Latha, K., Velraj, R., Shanmugam, P., Sivanesan, S.: Mixing strategies of high solids anaerobic co-digestion using food waste with sewage sludge for enhanced biogas production. J. Clean. Prod. **210**, 388–410 (2019)
23. Lee, J.H., Kim, D.H., Ryu, Y., Kim, K.H., Jeong, S.H., Kim, T.Y., Cha, S.W.: Mechanical properties of biocomposites using polypropylene and sesame oil cake. Polymers **13**(10), 1602 (2021)
24. Monlau, F., Latrille, E., Da Costa, A.C., Steyer, J.P., Carr`ere, H.: Enhancement of methane production from sunflower oil cakes by dilute acid pretreatment. Appl. Energy **102**, 1105–1113 (2013)
25. Nascimento, E.M.G.C., Carvalho, C.W.P., Takeiti, C.Y., Freitas, D.G.C., Ascheri, J.L.R.: Use of sesame oil cake (*Sesamum indicum L.*) on corn expanded extrudates. Food Res. Int. **45**(1), 434–443 (2012)
26. Pan, Y., Zhi, Z., Zhen, G., Lu, X., Bakonyi, P., Li, Y.Y., Zhao, Y., Banu, J.R.: Synergistic effect and biodegradation kinetics of sewage sludge and food waste mesophilic anaerobic co-digestion and the

underlying stimulation mechanisms. Fuel **253**, 40–49 (2019)

27. Paritosh, K., Yadav, M., Kesharwani, N., Pareek, N., Karthyikeyan, O.P., Balan, V., Vivekanand, V.: Strategies to improve solid state anaerobic bioconversion of lignocellulosic biomass: an overview. Bioresour Technol. **331**, 125036 (2021)
28. Pohl, M., Sanchez-Sanchez, M., Mumme, J.: Anaerobic digestion of wjeat straw and rape oil cake in a two-stage solid-state system. Renew. Energy **141**, 359–367 (2019)
29. Raposo, F., Borja, R., Martín, M., Martín, A., De la Rubia, M., Rincon´, B.: Influence of inoculum-substrate ratio on the anaerobic digestion of sunflower oil cake in batch mode: process stability and kinetic evaluation. Chem. Eng. J. **149**, 70–77 (2009)
30. Siddique, M.N.I., Wahid, Z.A.: Achievements and perspectives of anaerobic co-digestion: A review. J. Clean. Prod. **194**, 359–371 (2018)
31. Strazzera, G., Battista, F., Garcia, N.H., Frison, N., Bolzonella, D.: Volatil fatty acids production from food wastes for biorefinery platforms: A review. J. Environ. Manag **226**, 278–288 (2018)
32. Tang, J., Wang, X.C., Hu, Y., Pu, Y., Li, Y.: Nutrients removal performance and sludge properties using anaerobic fermentation slurry from food waste as an external carbon source for wastewater treatment. Bioresour Technol. **271**, 125–135 (2019)
33. Wang, Y., Li, G., Chi, M., Sun, Y., Zhang, J., Jiang, S.: Effects of co-digestion of cucumber residues to corn stover and pig manure ratio on methane production in solid state anaerobic digestion. Bioresour Technol. **250**, 328–336 (2018)
34. Yasothai, R.: Chemical composition of sesame oil cake-review. Int. J. Sci. Environ. **3**(3), 827–835 (2014)
35. Zhang, L., Loh, K.C., Zhang, J.: Enhanced biogas production from anaerobic digestion of solid organic wastes: Current status and prospects. Bioresour Technol. Reports **5**, 280–296 (2019)
36. Zhao, Y., Sun, F., Yu, J., Cai, Y., Luo, X., Cui, Z., Hu, Y., Wang, X.: Co-digestion of oat straw and cow manure during anaerobic digestion: Stimulative and inhibitory effects on fermentation. Bioresour Technol. **269**, 143–152 (2018)

Figures

Figure 1

Change of pH beside amount of VFAs

Figure 2

Removal of VS and TS in anaerobic co-digestion of FSOC and sewage sludge

Figure 3

Cumulative biogas, methane and carbon dioxide yields from anaerobic co-digestion of sewage and FSOC

Figure 4

Variation of specific biogas yield from anaerobic co-digestion of sewage and FSOC

Figure 5

CPI (co-digestion performance index) of different samples