Article

The Effects of Using Pretreated Cotton Gin Trash on the Production of Biogas from Anaerobic Co-Digestion with Cow Manure and Sludge

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Abstract: Anaerobic co-digestion (AcoD) has been practiced for decades to convert waste into value-added energy products, especially biogas. This study aimed to assess the potential of biogenic methane (CH₄) production from the co-digestion of pretreated cotton gin trash (CGT), cow manure, and sludge. CGT contains high cellulosic content, making it a reliable feedstock for biogenic methane production. To further improve the biogas quantity and quality, the CGT was subjected to physical pretreatments, i.e., hot water (HW), ultra-sonication (US), and a combination of both (HW+US). After 91 days of AcoD, 79–110 L of biogas was produced by the treatments. Among the treatments, HW+US-pretreated CGT presented maximum biogas production capacity, at 110 L. Besides, this treatment showed the high-quality biogenic CH₄ content, 52.4% of the total biogas volume, with an improved conversion rate of 0.37 L/g of volatile suspended solids consumed. In addition, this study discussed the structural changes in feedstock due to pretreatments and correlated them with the corresponding biogenic methane production. The study reports the potential of pretreated CGT conversion to CH₄. It will impact the circular economy by contributing to on-farm energy requirements and reducing the financial expenditures incurred in this regard.

Keywords: anaerobic co-digestion; alternative energy; biomethane; biogas; cotton gin trash; waste to energy

1. Introduction

Co-digestion is a well-established and conventional method where a combination of different feedstock is processed into the fundamental anaerobic digestion technique

to increase the biogenic methane content of produced biogas [1]. During the last two decades, a wide variety of biomass has been co-digested for bioenergy production. Among this common biomass, a few remarkable feedstocks have been reported as a promising output for methane production, such as cotton wastes [2], organic solid waste [3], sewage sludge [4], rice straw [5], and microalgae [6]. Lay et al. [7] reported that 1 g of sludge, meat, and vegetables produced 450, 424, and 203 to 269 mL of methane (CH_4), respectively. This result suggests that the plant-based recalcitrant material has less methane production capacity than the protein-based and stabilized biomass. This consequence can occur due to the high nitrogen content and resulting carbon to nitrogen ratios [7].

Compared with mono-digestion, co-digestion offers better nutrient availability, digestion stability, and degradation rate [8]. The co-substrates allow a better supply of nutrients and stability in biogas production. Gao et al. [9] reported that the biogenic methane production increased by 414% when the spent mushroom substrate was co-digested with chicken manure. This is due to the improved carbon to nitrogen (C:N) ratios in the co-biomass, which helps to better degrade the microorganisms. Similar results have been reported by Gonzalez-Piedra et al. [10], where raw cheese, whey, and coffee pulp were co-digested. Orangun et al. [11] compared the outcomes of mono- and co-digestion through laboratory experiments of goat manure and food waste. This experimental study determined that the co-digestion-obtained methane production capacity, 97.4%, presented better biodegradability than mono-digestion (33–65%). This degradation rate resulted in twice the amount of biogenic methane production.

A helpful energy material produced by anaerobic co-digestion is “biogenic methane”, a carbon-rich gas produced by microorganisms through biological reactions. The anaerobic digestion process is mainly governed by bacteria that convert carbon-rich biomass into different organic compounds. This leads to biogas production, almost 60% CH_4 , 40% carbon dioxide (CO_2), and few trace gases. Anaerobic digestion helps to reduce the emissions from biomass if disposed of directly and has the potential to produce CH_4 -rich biogas that can be used for energy production [8]. Moreover, it has the potential to produce comparatively stabilized manure that can be used as a soil conditioner due to the presence of rich nitrogen and phosphorus content [11].

Cotton waste is one of the most obnoxious wastes and is usually burnt on-farm to culminate insects and vectors, especially in continuous cotton cropping areas. This waste can be a compelling energy source, especially for countries with strict environmental regulations. Countries such as the United States, China, Brazil, India, Pakistan, Turkey, and Australia meet the world's cotton demand. Hence, this waste can be utilized as a biogas-producing raw material [12]. Isci and Demirer [12] reported 65 to 78 mL of CH_4 production from 1 g of cotton waste. An experimental study presented that cotton gin trash (CGT) is produced in high amounts as waste every year worldwide, which contains high lignocellulosic components (i.e., about 60%) [13]. Lignocellulose is a polysaccharide with predominantly cellulose, hemicelluloses, and lignin. Anaerobic bacteria can easily digest lignocellulose components to produce biogas [14]. On the other hand, cattle manure (CM) is another highly proven efficient methane-producing raw material because of its crucial contribution in maintaining the C:N ratio in the feedstock [15–17].

The significant fate of cotton crop residues and cow manure is mostly combustion, either for fuel purpose or the waste minimization process, which is hazardous for nearby residents' environment and public health. Therefore, anaerobic digestion can stabilize this biomass to avoid health risks and produce biofuels such as hydrogen, biomethane, methanol, or ethanol [8]. Optimization of thermal and biological processes can achieve better efficiency in getting desired products [18,19].

The anaerobic co-digestion process includes converting available volatile content in the biomass into biogenic methane. The reaction parameters are optimized for better quality and quantity of biogas generation. These parameters include several pretreatment parameters such as stirring intensity, physical and chemical characteristics of feedstock, temperature, pH, and substrate/inoculum (S/I) ratio. Feedstock type and pretreatment of

feedstock play a pivotal role in methane production efficiency and overall accumulation. Pretreatment helps in cell lysis and does not affect the recalcitrant chemical composition of the feedstock [20].

Generally, pretreatments can be biological, chemical, physical, and thermal [21]. Biological pretreatments are time-consuming and contain hypersensitivity issues. Therefore, most of the pretreatments only involve the addition of inoculum to the co-biomass. The chemical pretreatment (alkaline treatment) has been reported for high methane yield, as the addition of sodium hydroxide (NaOH) helps in the degradation of lignin and provides microbes fair access to the contents of feedstock [22]. The only limitation of using alkaline pretreatment is that NaOH boosts the toxicity levels in the byproducts [23].

In comparison, thermal pretreatment involves the thermal hydrolysis of organic compounds in feedstock [24]. This pretreatment improves the accessibility of microbes to volatile compounds. However, at high temperatures, the risks of low degradation of the solids increase [25]. Physical pretreatment includes mechanical means of cell lysis. These pretreatments mainly include ultra-sonication or centrifugation. Biogenic methane production has been reported to increase by 34% due to ultra-sonication (US) in pretreatment sludge [26].

The novelty of this study is to determine the effects of thermal and physical pretreatment on CGT for a better quality of biogas production co-digested with sludge and cow manure. To the authors' best knowledge, no earlier study has been demonstrated on this feedstock mixture for biogas production as well as biogas quality enhancement. Since all of these wastes are produced abundantly worldwide, quality biogas production from these feedstocks may boost the circular economy. This experimental study hypothesized that the pretreatments of feedstock (CGT) are not the only environment-friendly process. This method can also positively affect the efficiency of biogas/methane production. The study's main objective is to analyze the effect of ultra-sonication (US) and hot water (HW) pretreatment on CGT to produce biogenic methane. The study revealed the biogas production potential of pretreated CGT and the effects of pretreatment on biogenic methane production. Suppose that the pretreated CGT is successfully converted to biogenic methane; in that case, it will boost the circular economy, despite various issues [27,28], by contributing to on-farm energy requirements and soil conditioning [29], reducing the financial expenditures incurred in this regard. The study also discusses chemical changes in CGT structure after various pretreatments.

2. Materials and Methods

The digestate mixtures of CGT, CM, and sludge (as inoculum) were prepared for feeding into the reactor. The fresh samples of CGT, CM, and inoculum sludge were obtained from Varisco cotton gin near College Station, in Brazos County, Texas, animal science (ASTREC) facility at Texas A&M University, and the recycling stream of the anaerobic digester in the Texas A&M for sewage sludge, respectively. Four treatments were made, i.e., HW, US, HW+US, and no pretreatment (NP).

2.1. Pretreatment of CGT

The CGT was pretreated with three types of pretreatments (US, HW, and the combination of US and HW). The ultra-sonicator (Hielscher Ultra-sonic Processors, Ringwood, NJ, USA) was used to ultra-sonicate a 10% CGT solid solution for 30 min at the highest amplitude (100%) and cycle (1). An autoclave (Astell 130 L Top Loading Autoclave, NY, USA set at 121 °C and 15 psi) was used for the HW treatment of 10% solid solution in Borex bottles for one hour. Both pretreatments were combined after subjecting the samples to both methods.

2.2. Experimental Setup

Four lab-scale PVC batch digesters were used for this experiment. The construction of the layout is shown in Figure 1. The digesters were placed at room temperature, i.e.,

35 ± 2 °C. The mixture was made with CGT 52.27 g (by weight), one kg of CM, and one liter of inoculum sludge. The volume of gas produced was measured as per each gas collector's rise in water level. The reactors were inverted once a day to maintain the moisture content and maximum contact between substrates and microorganisms. The pH of the digesters was maintained by using a 5 M NaOH solution.

Figure 1. Layout of the experimental setup.

2.3. Analytical Methods

The effects of pretreatment on the structural composition of CGT were determined using Sluiter et al. [29]. The structural properties of CGT were observed using Fourier Transform InfraRed (FT-IR) spectroscopy, IR Affinity-1 with a MIRacle universal sampling accessory (Shimadzu, Kyoto, Japan) before and after pretreatments at the range of $4000\text{--}700\text{ cm}^{-1}$ (resolution 4 cm^{-1}).

The Proximate and Ultimate analysis of the three feedstocks was carried out. In proximate analysis, moisture content (MC), volatile combustible matter (VCM), and fixed carbon (FC) and ash content were determined by ASTM E871-72, ASTM E872-82, and ASTM D1102-84, respectively. The ultimate analysis of the biomass was carried out using Vario MICRO Elemental Analyzer, where carbon (C), hydrogen (H), sulfur (S), and nitrogen (N) were analyzed by ASTM E777, ASTM E775, and ASTM E778, respectively. The heating value of biomass was also tested using PARR isoperibol bomb calorimeter, IL, USA.

2.4. Gas Analysis

The gas sampling was carried out when the gas collectors were refilled (Table 1). The gas composition was analyzed by SRI Gas Chromatograph equipped with an on-column injection system and two detectors (Helium Ionization Detector and Thermal Conductivity Detector).

Table 1. Gas sampling intervals.

Day	Sampling Interval (Days)
1	
6	5
12	6
18	6
23	5
29	6
35	6
40	5
46	6
52	6
57	5
63	6
69	6
74	5
80	6
86	6
91	5

3. Results

3.1. Composition of Biomass

The CM and CGT have high carbon and volatile suspended solids (VSS) with the lowest sulfur content presented in Table 2. The sludge (inoculum) had high nitrogen content but minimal VSS. These results suggested that the sludge was stabilized, and it carried out the minimal microbial activity within its mass. The CGT can potentially serve as a significant source of nutrients and VSS for the AcoD process. Adding CM further improves nutrient availability and segregation. The inoculum to substrate ratio in VSS terms was 0.24. In comparison to mono-digestion, co-digestion provides enhanced digestibility due to the synergistic effects of the co-substrates, i.e., CM and CGT in this case [8]. This combination had the potential of producing high nutrient value of the products, which can play a positive impact on soil nourishment.

Table 2. Feedstock composition for anaerobic co-digestion.

Composition	Cotton Gin Trash	Cow Manure	Sludge
	Percentage Except for C/N and HHV		
Carbon	40.23	36.13	25.89
Nitrogen	2.36	2.3	5.97
Hydrogen	5.23	4.67	3.83
Sulfur	0.82	0.11	0.45
C/N	17.05	16.01	6.52
VSS (% of TS)	89.68	71.5	24.93
VCM	73.1	70.12	62.85
Fixed Carbon	16.7	12.1	22.74
Ash	10.2	17.1	14.4
High Heating Value (MJ/kg)	19.82	17.98	20.53

As suggested by Karki et al. [8], the process stability was attempted to be maintained by the pretreatment of CGT. The FTIR spectra of feedstock in Figure 2 showed visible and significant changes in the relative abundance of organic compounds for CGT. This can be observed in the pretreated US+HW. The ultra-sonication significantly changed CGT properties due to high volatile content. In addition, the ultra-sonication tended to break down the physical structure of CGT and resulted in an increased surface area, which may lead to solubilization and the further enhancement of the biogenic methane production [30].

Figure 2. Chemical structure of biogas produced from the digestion of cotton gin trash (FTIR spectra).

On the other hand, HW did not change the overall structure of CGT significantly. The use of acid or base as a pretreatment helped dissolve the lignocellulosic content and made it available for biological degradation, though it resulted from the minor intoxication of the byproducts. In the case of HW, a neutral solvent, partial dissolution was achieved along with low/no toxicity of the byproducts. Therefore, a combination of US+HW was expected to produce high-quality products at this stage.

Figure 2 shows that cellulose (C-O-C) had a strong peak in all the treatments in the fingerprint region of the spectra. The predominant species affected by the pretreatments include lignin, carboxylic acids, alcohols, and phenols. Peaks of organic species are well pronounced in US+HW-pretreated CGT. The relative abundance of organic species in the group frequency region of the spectra can be drawn as lignin (C-O-C) > aromatics (C-H) > alcohols and phenols (O-H) > carboxylic acids (C=O) > alkynes (C-C).

3.2. Biogas Production

The biogas production fluctuated during the digestion period, but the overall trend kept increasing consistently, especially for pretreated feedstock presented in Figure 3a. The fluctuations can be observed in untreated feedstock (i.e., no pretreatment) compared to pretreated feedstock runs, which show a comparatively uniform biogas production. These fluctuations have already been reported by Twizerimana et al. [2], as the highest production rates were observed on day 21, when cotton yarn waste was digested. These fluctuations can be attributed to the type and condition of the biomass used for the conversion process.

Saleem et al. [6] has reported that the hot water and ultra-sonication pretreatment decreased the methane production for microalgae. Figure 3b shows the cumulative biogas production for 91 days, where the total output of biogas among treatments remained as US+HW > HW > NP > US pretreatments. This suggested that the aggregation of bulky feedstock components by ultra-sonication helps to produce high amounts of biogas when dissolved in the solvent, i.e., HW in the current study. This experimental output represented that the type and composition of biomass is a significant attribute to be considered for selecting the pretreatments.

Figure 3. Biogas yield of the co-digestion of cotton gin trash with different pretreatments. Actual biogas generation: liters per sampling interval, (a), cumulative biogas production; liters, (b), biogas production; liters per day, (c), and average biogas production rate in three phases; liters per day, (d).

If the biogas production rates (liters/day) are compared graphically, the overall biogas production can be divided into three phases, which are presented in Figure 3c. In the initial phase, acclimatization occurs, meaning the biogas production rates are at the minimum, constantly increasing in the second phase. The biogas production then comparatively stabilizes in the third phase. In Figure 3c, it can be observed that the biogas production rates for all the treatments remained almost similar, whereas, in the subsequent two steps, the production was changed with all four treatments. The production rates were divided into the corresponding three phases based on the trends. Although it depends on the type of biomass, phases can be generalized as phase 1, phase 2, and phase 3 from day 1 to 36, day 37 to 60, and day 60 to higher, respectively.

Figure 3d compares the average biogas production rates of four treatments. The average biogas production rates for all the treatments were significantly diverse in all three phases except for ultra-sonication pretreatment. The feedstock pretreated with ultra-sonication increased the biogas production in the second phase but could not maintain the same in the third phase. This is probably due to the lesser availability of feedstock components for degradation than the feedstock treated with HW. It can be concluded that ultra-sonication can help in the rapid increase in biogas production, but to continue the production, a better solvent is required, which is HW in our case.

The digestion period and biogas production have been normalized to observe the gas production rate. Figure 4 shows that, initially, the biogas production was increased. Still, it ceased for a few days in both treated and untreated CGT treatments, which showed the rapid digestion of simple saccharides and carbohydrates. Then, the further increase in the gas production rate can be attributed to the degradation of complex recalcitrant organic material. The overall trend for the digestion process remained almost similar for pretreated and non-pretreated CGT.

Figure 4. Normalized biogas production rate.

For theoretical calculations of biogas production, the curves were fitted by a power function (i.e., coefficient of biogas production) followed by Equations (1) and (2).

$$F(X) = X^p \quad (1)$$

$$\ln(F(X)) = p \ln(X) \quad (2)$$

where, p = coefficient of biogas production; X = cumulative gas production/total gas production; $F(X)$ = cumulative time/total time.

It is pertinent to mention here that if the coefficient of biogas production for treatments is higher than 1 (i.e., 45° line), it shows significant biogas production in a limited time. The non-pretreated CGT showed a consistent biogas production, as the p -value was almost 1 (Figure 4d). The hot water and ultra-sonication showed a higher biogas production rate (Figure 4a), and the hot water and ultra-sonication decreased the biogas production (Figure 4b,c). It can be concluded that the pretreatments significantly affected the biogas production and combined treatment of hot water and ultra-sonication showed positive results.

3.3. Biogas Composition

Table 3 shows the quality of biogas produced and process efficiency. As biogenic methane production was the ultimate goal of the anaerobic digestion process, the volume of methane gas in the biogas produced by all treatments is discussed in Table 2. It is essential to mention here that the quality of biogas production is much more important than quantity. The ultra-sonication-pretreated CGT produced the lowest biogas, but the methane content knocked out the untreated feedstock. The combined pretreated (US+HW) feedstock not only produced high amounts of biogas, but also had high biogenic methane content (52%). This helps in deducing that pretreatments can play a pivotal role in producing better quality products.

Table 3. Production of methane by each treatment with different pretreatments.

Pretreatment	VSS _{consumed}	Biogas _{produced}		CH ₄ _{produced}	
	(g)	(L)	(% of Biogas)	(L)	(L/g of VSS)
HW	138.7	93.17	37.83	35.25	0.25
US	129.5	79.66	37.58	29.94	0.23
HW+US	145.2	103.99	52.4	54.49	0.37
NP	131.7	87.685	31.6	27.71	0.21

The process efficiency is shown in Table 2 by comparing the total amount of biogenic methane with the volatile suspended solids consumed. Based on this parameter, the most

efficient conversion took place for the CGT treated with both US+HW (0.37 L/g of VSS), i.e., almost 0.6 times higher than the other treatments, and the least efficient conversion remained for the untreated CGT (NP = 0.21 L/g of VSS).

The stoichiometric theoretical yields were calculated by converting the results of the elemental analysis of the feedstock and their respective weights used in the treatments [31]. The molar weights were then used to balance the stoichiometric conversion arithmetically. The total theoretical yield calculated for our treatments was 138 L of methane. Compared with the stoichiometric theoretical yield [31], the US+HW-treated CGT produced almost 40% of the theoretical yield (4.79 mol CH₄). The other three treatments yielded 22% of the theoretical yield on average. This suggests that other gases such as CO₂, CO, NH₃, H₂S, etc., were produced in higher quantity than CH₄, demonstrating that the pretreatment of US+HW can improve the process efficiency.

3.4. Substrate Composition

The composition showed that carbon constitutes a significant proportion of the leftover substrate (Figure 4). Generally, the average C:N increased in the substrate with time, suggesting the depletion of the nitrogen species in the substrate. Specifically, the trend kept increasing over time, except for the CGT treated with HW. The C:N increased to 25 at day 45 in this treatment and declined to 20 on day 91. This trend can be correlated with high microbial activity during the second digestion phase (presented in Figure 5).

Figure 5. Ultimate substrate analysis during the digestion process of cotton gin trash.

The relative percentage of C, H, N, and S remained consistent in CGT treated with US+HW. This consistency can be correlated with the biogas production rate, which showed an exponentially increasing trend. The combination of US+HW might have improved the bioavailability or inoculum–substrate interaction, resulting in continuous and consistent biogas production. This results in the timely occurrence of different phases of the co-digestion process, i.e., hydrolysis, acido-, acito-, and methanogenesis [32,33].

Although the pretreatment of feedstock always increases the processing costs [27], the budgeting of the complete process can help understand the applicability of such techniques. Scherzinger and Kaltschmitt [34] reported that the pretreatments could reduce the overall cost for power generation as they can increase the valued products. Each pretreatment has its own economic and environmental pros and cons [35]. Life cycle assessments (LCA) can be conducted to evaluate the impact of these pretreatments on the environment. It

is essential to consider the transportation/handling costs, disposal costs, environmental degradation costs, and market value of the products. When all these factors are considered, it is hypothesized that the overall conversion of wastes to valuable products can become feasible both economically and environmentally [35]. A further study on all the economic factors can help implement our results on a commercial scale.

4. Conclusions

The process of anaerobic co-digestion (AcoD) is paving the way for alternate energy resources. Process optimization and feedstock iterations are helping in achieving target energy products. We evaluated the potential of biogenic methane production from CGT with process optimization using pretreatments (US+HW). Almost 79 to 110 L of biogas were produced in 91 days of the digestion. The pretreatments significantly improved the quantity and quality of biogas. Among the treatments, the US+HW-combined-pretreated CGT had the highest biogas (110 L), with an improved biogenic methane content of 52.4% of the total biogas volume. The process efficiency for this treatment was also better than the other treatments at a conversion rate of 0.37 L of CH₄/grams of volatile suspended solids consumed. The core conclusions drawn from the co-digestion of CGT and CM can be summarized as:

1. The pretreatments significantly affected the chemical structure of CGT;
2. Pretreatments of cotton gin trash increased biogas production;
3. Among the pretreatments, hot water and ultra-sonication combined showed high biogas production and biogenic methane content;
4. The chemical structure remained consistent during the AcoD process;

Other wastes from cotton fields can also be co-digested to obtain a better quantity and quality of biogenic methane. It will also be helpful in decision-making regarding whether the economic feasibility of biogenic methane production is carried out to pinpoint the research directions further.

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References

1. Johnson, S.; Echeverria, D.; Venditti, R.; Jameel, H.; Yao, Y. Supply Chain of Waste Cotton Recycling and Reuse: A Review. *AATCC J. Res.* **2020**, *7*, 19–31. [[CrossRef](#)]
2. Twizerimana, M.; Marimi, M.; Bura, X.; Nganyi, E.O. Biogas Production from Co-digestion of Cotton Yarn Waste and Human Urine. *J. Energy Res. Rev.* **2020**, *6*, 20–29. [[CrossRef](#)]
3. Kumar, A.; Samadder, S. Performance evaluation of anaerobic digestion technology for energy recovery from organic fraction of municipal solid waste: A review. *Energy* **2020**, *197*, 117253. [[CrossRef](#)]

4. Hanif, M.U.; Zwawi, M.; Capareda, S.C.; Iqbal, H.; Algarni, M.; Felemban, B.F.; Bahadar, A.; Waqas, A. Influence of Pyrolysis Temperature on Product Distribution and Characteristics of Anaerobic Sludge. *Energies* **2019**, *13*, 79. [\[CrossRef\]](#)
5. Luo, T.; Khoshnevisan, B.; Pan, J.; Ge, Y.; Mei, Z.; Xue, J.; Fu, Y.; Liu, H. How exothermic characteristics of rice straw during anaerobic digestion affects net energy production. *Energy* **2020**, *212*, 118772. [\[CrossRef\]](#)
6. Saleem, M.; Hanif, M.U.; Bahadar, A.; Iqbal, H.; Capareda, S.C.; Waqas, A. The Effects of Hot Water and Ultrasonication Pretreatment of Microalgae (*Nannochloropsis oculata*) on Biogas Production in Anaerobic Co-Digestion with Cow Manure. *Processes* **2020**, *8*, 1558. [\[CrossRef\]](#)
7. Lay, J.J.; Li, Y.Y.; Noike, T.; Endo, J.; Ishimoto, S. Analysis of environmental factors affecting methane production from high-solids organic waste. *Water Sci. Technol.* **1997**, *36*, 493–500. [\[CrossRef\]](#)
8. Karki, R.; Chuenchart, W.; Surendra, K.; Shrestha, S.; Raskin, L.; Sung, S.; Hashimoto, A.; Khanal, S.K. Anaerobic co-digestion: Current status and perspectives. *Bioresour. Technol.* **2021**, *330*, 125001. [\[CrossRef\]](#)
9. Gao, X.; Tang, X.; Zhao, K.; Balan, V.; Zhu, Q. Biogas Production from Anaerobic Co-Digestion of Spent Mushroom Substrate with Different Livestock Manure. *Energies* **2021**, *14*, 570. [\[CrossRef\]](#)
10. Gonzalez-Piedra, S.; Hernández-García, H.; Perez-Morales, J.; Acosta-Domínguez, L.; Bastidas-Oyanedel, J.-R.; Hernandez-Martinez, E. A Study on the Feasibility of Anaerobic Co-Digestion of Raw Cheese Whey with Coffee Pulp Residues. *Energies* **2021**, *14*, 3611. [\[CrossRef\]](#)
11. Orangun, A.; Kaur, H.; Kommalapati, R. Batch Anaerobic Co-Digestion and Biochemical Methane Potential Analysis of Goat Manure and Food Waste. *Energies* **2021**, *14*, 1952. [\[CrossRef\]](#)
12. Isci, A.; Demirel, G. Biogas production potential from cotton wastes. *Renew. Energy* **2007**, *32*, 750–757. [\[CrossRef\]](#)
13. Haque, A.N.M.A.; Remadevi, R.; Naebe, M. A review on cotton gin trash: Sustainable commodity for material fabrication. *J. Clean. Prod.* **2020**, *281*, 125300. [\[CrossRef\]](#)
14. Plácido, J.; Imam, T.; Capareda, S. Evaluation of ligninolytic enzymes, ultrasonication and liquid hot water as pretreatments for bioethanol production from cotton gin trash. *Bioresour. Technol.* **2013**, *139*, 203–208. [\[CrossRef\]](#)
15. Allah, W.A.; Tawfik, M.; Sagade, A.A.; Gorjian, S.; Metwally, K.; El-Shal, H. Methane production enhancement of a family-scale biogas digester using cattle manure and corn stover under cold climates. *Sustain. Energy Technol. Assess.* **2021**, *45*, 101163. [\[CrossRef\]](#)
16. Wang, K.; Yun, S.; Xing, T.; Li, B.; Abbas, Y.; Liu, X. Binary and ternary trace elements to enhance anaerobic digestion of cattle manure: Focusing on kinetic models for biogas production and digestate utilization. *Bioresour. Technol.* **2020**, *323*, 124571. [\[CrossRef\]](#)
17. Ma, G.; Ndegwa, P.; Harrison, J.H.; Chen, Y. Methane yields during anaerobic co-digestion of animal manure with other feedstocks: A meta-analysis. *Sci. Total Environ.* **2020**, *728*, 138224. [\[CrossRef\]](#)
18. Prabhu, A.V.; Raja, S.A.; Avinash, A.; Pugazhendhi, A. Parametric optimization of biogas potential in anaerobic co-digestion of biomass wastes. *Fuel* **2020**, *288*, 119574. [\[CrossRef\]](#)
19. Mirmohamadsadeghi, S.; Karimi, K.; Azarbaijani, R.; Yeganeh, L.P.; Angelidaki, I.; Nizami, A.-S.; Bhat, R.; Dashora, K.; Vijay, V.K.; Aghbashlo, M.; et al. Pretreatment of lignocelluloses for enhanced biogas production: A review on influencing mechanisms and the importance of microbial diversity. *Renew. Sustain. Energy Rev.* **2020**, *135*, 110173. [\[CrossRef\]](#)
20. Tiehm, A.; Nickel, K.; Zellhorn, M.; Neis, U. Ultrasonic waste activated sludge disintegration for improving anaerobic stabilization. *Water Res.* **1997**, *35*, 2003–2009. [\[CrossRef\]](#)
21. Luna-Avelar, K.D.; Barrena, R.; Font, X.; Sánchez, A.; Santos-Ballardo, D.U.; Germán-Báez, L.J.; Valdez-Ortiz, A. A preliminary assessment of anaerobic co-digestion potential of mango and microalgal residue biomass using a design of experiments approach: Effect of thermal, physical and biological pretreatments. *Food Bioprod. Process.* **2021**, *128*, 143–152. [\[CrossRef\]](#)
22. Gunaseelan, V.N. Anaerobic digestion of biomass for methane production: A review. *Biomass Bioenergy* **1997**, *13*, 83–114. [\[CrossRef\]](#)
23. Kim, J.S.; Lee, Y.Y.; Kim, T.H. A review on alkaline pretreatment technology for bioconversion of lignocellulosic biomass. *Bioresour. Technol.* **2016**, *199*, 42–48. [\[CrossRef\]](#)
24. Rajput, A.A.; Zeshan; Hassan, M. Enhancing biogas production through co-digestion and thermal pretreatment of wheat straw and sunflower meal. *Renew. Energy* **2021**, *168*, 1–10. [\[CrossRef\]](#)
25. Mladenovska, Z.; Hartmann, H.; Kvist, T.; Sales-Cruz, M.; Gani, R.; Ahring, B.K. Thermal pretreatment of the solid fraction of manure: Impact on the biogas reactor performance and microbial community. *Water Sci. Technol.* **2006**, *53*, 59–67. [\[CrossRef\]](#)
26. Kim, D.-H.; Jeong, E.; Oh, S.-E.; Shin, H.-S. Combined (alkaline+ultrasonic) pretreatment effect on sewage sludge disintegration. *Water Res.* **2010**, *44*, 3093–3100. [\[CrossRef\]](#)
27. González-Arias, J.; Baena-Moreno, F.M.; Gonzalez-Castaño, M.; Arellano-García, H.; Lichtfouse, E.; Zhang, Z. Unprofitability of small biogas plants without subsidies in the Brandenburg region. *Environ. Chem. Lett.* **2021**, *19*, 1823–1829. [\[CrossRef\]](#)
28. González-Arias, J.; Baena-Moreno, F.M.; Pastor-Pérez, L.; Sebastia-Saez, D.; Fernández, L.M.G.; Reina, T. Biogas upgrading to biomethane as a local source of renewable energy to power light marine transport: Profitability analysis for the county of Cornwall. *Waste Manag.* **2021**, *137*, 81–88. [\[CrossRef\]](#)
29. Bonetta, S.; Bonetta, S.; Ferretti, E.; Fezia, G.; Gilli, G.; Carraro, E. Agricultural Reuse of the Digestate from Anaerobic Co-Digestion of Organic Waste: Microbiological Contamination, Metal Hazards and Fertilizing Performance. *Water Air Soil Pollut.* **2014**, *225*, 2046. [\[CrossRef\]](#)

30. Sluiter, A.; Hames, B.; Hyman, D.; Payne, C.; Ruiz, R.; Scarlata, C.; Sluiter, J.; Templeton, D.; Wolfe, J. Determination of Total Solids in Biomass and Total Dissolved Solids in Liquid Process Samples, Laboratory Analytical Procedure. Technical Report NREL/TP-510-42621, National Renewable Energy Laboratory 2008. Available online: <https://www.nrel.gov/docs/gen/fy08/42621.pdf> (accessed on 11 December 2021).
31. Qi, N.; Zhao, X.; Zhang, L.; Gao, M.; Yu, N.; Liu, Y. Performance assessment on anaerobic co-digestion of Cannabis ruderalis and blackwater: Ultrasonic pretreatment and kinetic analysis. *Resour. Conserv. Recycl.* **2021**, *169*, 105506. [[CrossRef](#)]
32. Buswell, A.M.; Mueller, H.F. Mechanism of Methane Fermentation. *Ind. Eng. Chem.* **1952**, *44*, 550–552. [[CrossRef](#)]
33. Meegoda, J.N.; Li, B.S.-K.; Patel, K.; Wang, L.B. A Review of the Processes, Parameters, and Optimization of Anaerobic Digestion. *Int. J. Environ. Res. Public Health* **2018**, *15*, 2224. [[CrossRef](#)]
34. Scherzinger, M.; Kaltschmitt, M. Techno-economic assessment of common reed and sewage sludge co-fermentation improved by vapothermal pre-treatment. *Clean Technol. Environ. Policy* **2021**, *23*, 2741–2755. [[CrossRef](#)]
35. Mainardis, M.; Buttazzoni, M.; Gievers, F.; Vance, C.; Magnolo, F.; Murphy, F.; Goi, D. Life cycle assessment of sewage sludge pretreatment for biogas production: From laboratory tests to full-scale applicability. *J. Clean. Prod.* **2021**, *322*, 129056. [[CrossRef](#)]