

# Ultrasonic Pre-treatment Investigation on Biogas Production of Biomethane of Fruit and Vegetable Waste

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Structural and compositional characteristics of substrate in anaerobic digestion are known to limit the hydrolysis step. This can be countered by the use of pretreatment prior to the commencement of anaerobic digestion process. In this instance pretreatment step assists in the increase of the degradability of the substrate. Thus accelerating hydrolysis and improving methane yields Ultrasonic pretreatment is a type of mechanical treatment mainly employed for surface area increase through solid particles disintegration. This is achieved by the use of grinding methods. In this work, the main objective was to investigate how ultrasonic pretreatment affects the biogas yield. Three sonication times of (10, 20, 45 min), operated at 20 MHz and an amplitude of 8  $\mu\text{m}$  were used on fruit and vegetable waste inoculated with sludge. BMP tests were performed on ultrasonic pre-treated substrate, with total solids (%TS) of 17.9%, volatile solids (%VS) of 82.1 %, moisture content of 82.1 % and C/N of 26%. The tests were performed in batch reactors, at mesophilic temperature of 35 °C. In this work, the batch time of 13 days was sufficient to complete the process of digestion. The highest cumulative methane production was 238  $\pm$ 7.64 mL/g VS, which occurred at 45 min sonication time. This yield is 43 % higher than the untreated sample. 20 minutes for pretreatment sonication time, resulted in the cumulative methane production of 210.4  $\pm$ 3.82 mL/g VS. Finally, sonication time of 10 minutes resulted in the least methane yield of 173  $\pm$ 3.18 mL/g VS, with only 22 % increase in the yield. These results led to the conclusion that in this instance, an increase in ultrasonic pretreatment time, resulted in an increased the methane yield.

## 1. Introduction

Increase rate in population growth as well national industrialization has led in increased consumption of energy and production of waste. Environmental issues such as pollution, global warming, acid rain, damage of the ozone layer are caused by inadequate and ineffective management of solid waste. The disposal of waste in South African landfills is a norm due to most of landfill sites' tipping fees being relatively low as compared to developed countries. However, landfilling has negative impacts on the environment due the production of landfill gas and leachates. Thus waste can be diverted to the developing energy sector as the ambition of the South African government green the economy. In South Africa, there is an opportunity for using fruit and vegetable waste as an energy source since energy recovery from waste is leaning more towards the direction of anaerobic digestion (AD), refuse-derived fuel processes and composting (Zeynali et al., 2017; Hartmann and Ahring, 2006; Notten et al., 2014). AD is one of the waste to energy (WtE) technologies that can produce fuels to be utilised in the production of electrical and or thermal energy, in vehicles fuel production and can also be injected into natural gas grid (Maisaraha et al., 2018).

### 1.1 Anaerobic digestion

Anaerobic digestion is the production of simpler compounds through the degradation of complex organic matter. This process occurs with the use of numerous microbial consortia which transform complex macromolecules into lower molecular weight compounds in the absence of oxygen with the. Methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>) with variable amounts of hydrogen sulphide (H<sub>2</sub>S), oxygen and water are the products of this process.

There are four biochemical steps contained in the anaerobic digestion process. The first step is called hydrolysis -hydrolytic bacteria remove polymers to monomers. Acidogenesis is the second step - acidogenic bacteria convert hydrolysis products into carboxylic acid, carbon dioxide, hydrogen and alcohol. A diverse bacterial community can ferment a simple substrate to produce different kinds of products, i.e. glucose degrading to acetate, ethanol and propionate. This is considered to be the most rapid step in anaerobic digestion. However, methanogenic bacteria can not directly use the products, hence a third step further degrades them (Chernicharo, 2007); Kangle, et al., 2012; Lier et al., 2008, Rea, 2014). The third step is called acetogenesis, in which the acidogenesis products are converted into acetic acid, hydrogen and carbon dioxide. )Finally, the last step is called methanogenesis –where bacteria called methanogens break down the acetic acid to produce methane (Van Fan et al., 2018, Adebayo et al., 2014, Krishna, 2013, Rea, 2014, Mudhoo and Kumar, 2013, Chernicharo, 2007, Kangle et al., 2012).

## **1.2. Pre-treatments**

Hydrolysis in AD can be limited by structural and compositional characteristics of substrate. For instance, uncooked vegetables contain starch, which has a higher degree of crystallinity and this is bound to hinder the hydrolysis step. This leads to the requirement of the pretreatment step to increase the degradability of the substrate. Pretreatment is not only known for the increase in biogas yields but also for the improvement in reduction of sludge volumes as well as organic matter. Pretreatment methods that can be employed prior to AD and methane yields are thermal, mechanical, chemical, and biological (Karlsson et al., 2011; Paritosh et al., 2017).

### **1.2.1 Physical pretreatment**

Mechanical and thermal treatment are types of physical treatments. Mechanical treatment is primarily used for increasing the surface area by disintegrating solid particles with the use of a grinding methods. Digestion is enhanced by the increase in surface area since the interaction between the substrate and inoculum is improved (Rena et al., 2018). Ultrasonication is a form of a mechanical pretreatment method. Cavitation bubbles are used to generate large hydro-mechanical shear force, this is achieved by use of high intensity ultrasonic waves, which in return disintegration of sludge. Ultrasonic method increases insoluble microbial product due to solubilization of both intracellular and extracellular substance and this method is known to be faster than the thermal pretreatment. In a study by Bani et al, 2016, thermal pretreatment was conducted on inoculums at 120 °C, 160 °C and 180 ° C for 2 hours. In this instance 160 °C and 180 °C, resulted in the biogas production improving by 0.8 times that of the untreated samples. A study conducted by Izumi et al. (2010), particle size reduction from 0.843 mm to 0.39 mm, resulted in a 28% increase in methane yield.

### **1.2.2 Biological pretreatment**

Biological pretreatment methods are known to require less costs in terms of capital, energy consumption, operations and maintenance. This type of treatment's research area is gaining much interest. (Kavitha et al., 2017; Rena, et al., 2018).

### **1.2.3 Chemical pretreatment**

Acid, alkaline and oxidation are types of chemical pretreatment. Acid pretreatment leads to the hydrolysis of carbohydrates, while alkaline and oxidation pretreatment attack the lignin. Alkaline pretreatment cause disrupts the structural connections between lignins and carbohydrates. This type of pretreatment increase internal surface area of lignocellulose, thus converting lignin into VFA, a substrate suitable for the production of biogas (Wang, 2011).There is a limited research on the effects of ultrasonic on the anaerobic digestion of solid waste, even though there is much research work done on the municipal waste water. Thus the main objective of this work is to investigate how ultrasonic pretreatment affects the biogas yield, in a batch bioreactor under mesophilic conditions.

## **2. Materials and method**

### **2.1 Materials**

The substrate used in this study was a mixture of FVW<sup>o</sup>, which was inoculated with sludge and a series of BMP (biomethane potential) tests were performed. Prior to the BPM tests, the substrate was subjected to an ultrasonic pretreatment

### **2.2 Sample collection**

Fruit and vegetable waste was collected from a local fruit and vegetables wholesaler. A 20 L container was used to collect samples at different locations to get well represented sample over a period of a week. The waste was sorted to remove unwanted material such as packaging material. While the sludge was collected from a waste water treatment plant's anaerobic digester at ERWAT (East Rand Water Care Company) in Kliprivier, South Africa.

## 2.3 Sample Preparation

To homogenize the FVW sample for the ultrasonic pretreatment as well as the BMP tests, an electric blender was utilized. When using the blender, no additional water was added to preserve the characteristics of the sample. Unused samples were stored in clearly marked plastics and stored in the refrigerator at -20 °C.

## 2.4 Ultrasonic pretreatment

The pretreatment in the form of ultrasonic was performed by Misonix Fischer Scientific Ultrasonic Sonicator XL2020, with 1, 5 inch diameter probe. It was operated at 20 MHz and an amplitude of 8  $\mu\text{m}$ . Different sonication times were 10, 20 and 45 minutes.

## 2.5 Analytical Methods for Organic Substrate Characteristics

All analytical determinations were conducted in accordance with Standard Methods of examination of water and waste water as prescribed by APHA (2005).

## 2.6 Methane production assays

Automatic methane potential test system (AMPTS II), Figure 1 (Bioprocess Control, Sweden, Ab) was used to conduct biomethane tests.

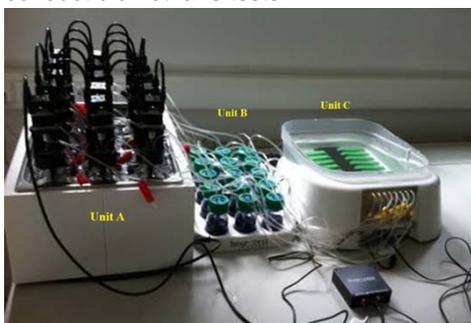


Figure. 1: Automatic methane potential test system (AMPTSII) (Browne, 2013)

The production rate of biogas was determined by feeding bioreactors with a mixture of FVW and the inoculum. The feed constituted of 80% water and 20 % of a mixture of FVW and inoculum. This resulted in the feed of 5% total solids (TS). It is important to expel oxygen by flushing the reactors with nitrogen ( $\text{N}_2$ ) before starting the experiments. This is done to make the environment conducive for anaerobic digestion. Hot water bath incubation unit was kept at 35 °C to maintain a constant mesophilic temperature for the experiment. Reactors are immersed in this water bath and make up the whole unit. A  $\text{CO}_2$ -fixing unit (unit B) which uses hydroxides, is used to trap gases such as hydrogen sulphide such and carbon dioxide. Agitation of 1min on/off was set to ensure that there was an efficient contact between the microbes and nutrients. Gas volume measuring device (unit C), measure the resulting biomethane via liquid displacement method. The results were recorded by an integrated data acquisition system. BMP tests were done in triplication. These tests were ran for 30 days.

## 3. Results an Discussions

### 3.1 Substrate characterization

Each fraction of fruit (peaches, bananas, nectarines and apples) and vegetable (butternuts, potatoes and carrots) were not quantitatively classified. The characteristics of the feedstock affects the anaerobic digestion process. The characteristics of the sampled FVW is presented in Table 1

Table 1: FVW characterization analysis

FVW	%
Fruits	60
Vegetables	40
TS	17.9
MC	82.1
VS	82.1
C/N	26

From Table 1, it can be observed that the substrate's total solids was 17%, which is in the range of the TS for food waste used in the study by Cho et al., (1995), which was between 15-30%. The higher solid contents of a substrate hinders the efficacy of bioconversion due to inadequate mixing as well as dead zones' existence within the bioreactor. Whereas lower solid content results in decreased availability of the substrate. Therefore the desirable range 10-25 % (Matheri et al., 2016) and the FVW % TS falls within that range. The moisture content and C/N ratio of the substrate was 82% and 26 %, respectively. Both which are comparable to the study by Vallejo et al, 2020 where it was reported that moisture content and C/N ratio of the substrate were 83% and 24 %, respectively. Generally, the feed's carbon content should be 25 – 30 times more than nitrogen since it was observed that microorganism's rate of carbon consumption is 25-30 times faster than nitrogen. Of high importance is that nitrogen rich substrates, i.e. substrate with low C: N ratio may cause ammonium inhibition, which is toxic to mesophilic methanogenic bacteria. Whilst, substrates with C: N ratio above 30 result in low protein formation due to multiplication of microorganism slowing down (Mahanta et al., 2005; Maile et al., 2016)

### 3.2 Methane production

As stated by Labatut et al., 2011 that factors such as production of inhibitory intermediate fermenters, biodegradability characteristics as well as methanogenic bacterial population's performance control the ultimate shape of the methane production curve. Figure 2 (a, b, c and d) show the cumulative methane production curves of fruit and vegetables waste.

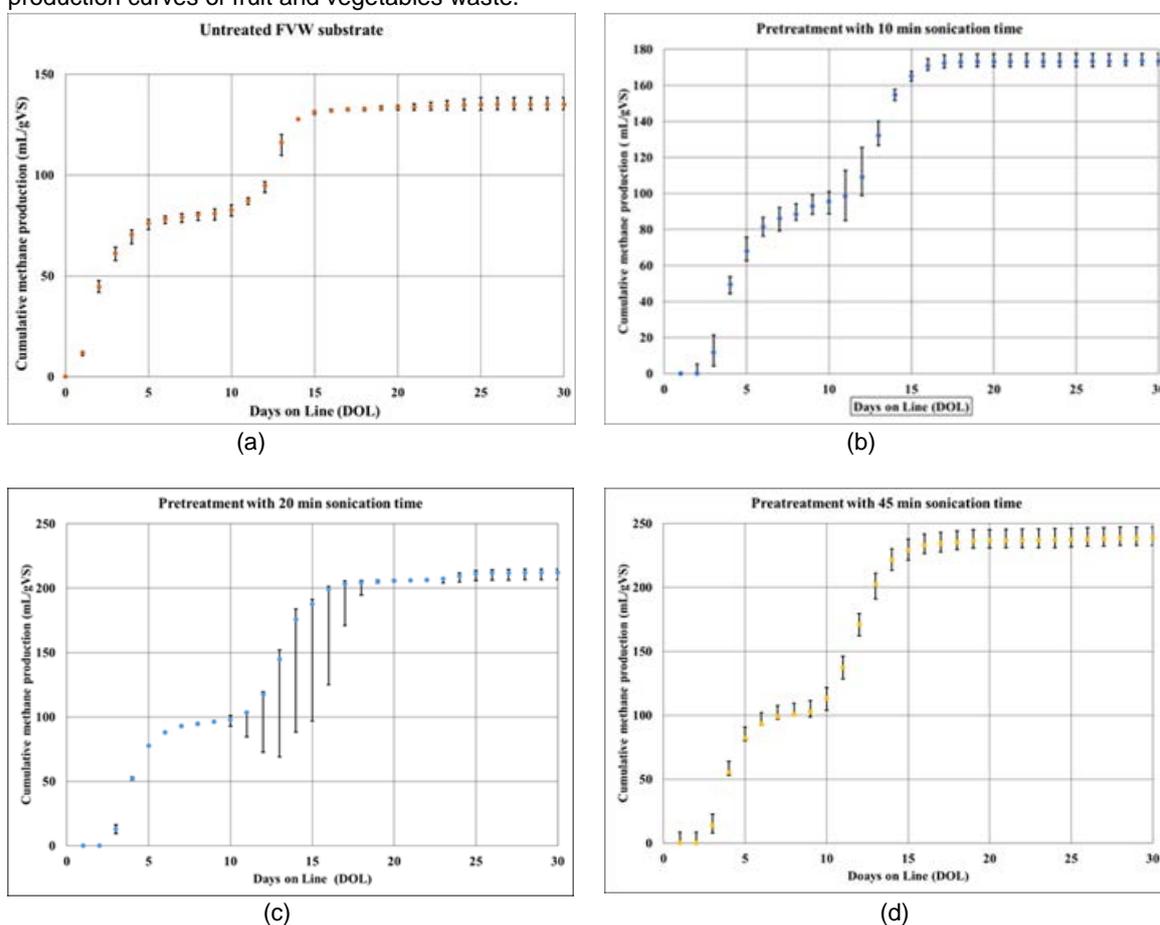


Figure 2: Cumulative methane production of untreated and ultrasonic pre-treated FVW

From Figure 2 (a) it is observed that from day 1 to 5, there was a steep increase in methane yield, which was then followed by a steady decrease in the methane production from day 5 to day 12. However this decrease was followed by another sharp increase of the methane yield from day 14 to 17. The first increase can be attributed to FVW containing easily hydrolysed organic matter such as sugars, whereas the decrease can be due to fibres which biodegrade slowly. The resulting average cumulative methane production was  $135.14 \pm 3.18$  mL/gVS. In Figure 2 (b) for 10 minutes sonication time pretreatment, it can be observed that from

day 1 to 5, there was a steep increase in methane yield, which was then followed by a steady decrease in the methane production from day 5 to day 10. However this decrease was followed by another sharp increase of the methane yield from day 10 to 15. The resulting average cumulative methane production was  $173.23 \pm 3.82$  mL/gVS, which is 22 % increase as compared to the untreated substrate. From Figure 2 (c), the second replicate shows larger error bars as this experiment differs from the other two. This experiment showed a much longer lag which extended from days 9 to 14. However this experiment was still used in this study, as it might provide crucial information about the system. The reason for this deviation might be a blockage in the system or inhibition. Labatut et al., 2011 stated that in some BMP assay can be affected by product inhibition, of which is difficult to prevent and affects the reaction kinetics. However, inhibition is reversible, provided that sufficient digestion time is allowed and thermodynamic conditions are favourable, the expected maximum methane yields can be achieved. This is shown in this instance, where the system recovered from it, as methane continued to be produced, even after the setback. The average cumulative methane produced was  $210.4 \pm 4.2$  mL/g VS which was 35 % more than the untreated substrate's yield. From Figure 2 (d), it can be observed that the average methane production of  $238.6 \pm 7.64$  mL/gVS was achieved and about 13 days was sufficient the digestion.

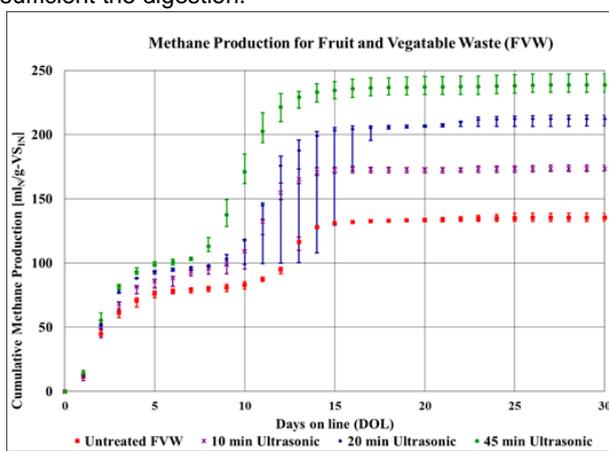


Figure 3: Cumulative methane production of FVW at different sonication times

From Figure 3, it was observed that FVW might have had a high lignin content, which takes a while to digest since it is hard to break down. It can be observed that their shapes are similar, with two distinct plateaus. The first plateau region represents the easily biodegradable matter, such as readily available sugars, which were hydrolysed easily and much quicker. Whereas the second plateau represents cellulose and starch which requires much time to breakdown and produce methane. This shows that in the initial step, the overall kinetics are faster due to the readily biodegradable organic matter being converted into biogas. After a certain time, with the remainder of slowly biodegradable matter, the rate slows down. As stated by Zeynali et al. (2017) that shorter batch time leads to an increase in profits due to smaller anaerobic digester or more feedstock loading rate. At 45 min sonication time, the effects of ultrasonic are observed with highest methane yield of 238 mL/g VS, which is 43 % higher than the untreated sample. The least methane yield of 173 mL/g VS occurred at sonication time of 10 minutes. This had only 22 % increase in the yield as compared to the untreated sample. In this study an increase in sonication time resulted in the increase in methane yield. In a study by Vieitez and Ghosh (1999), where the effects of ultrasonic pre-treatment on biogas yield from kitchen waste was investigated, it was found that the sonication time and density ( $W\ mL^{-1}$ ) greatly affects the yield of biogas. Finally, Zeynali et al. (2017) stated that the increase in the biogas yields might be due to the reduction in particle size as well as organic matter solubility improvement as a result of ultrasonic pre-treatment. (Srisowmeya et al., 2019) stated that due to the nature of the ultrasonication procedure, the solubility and accessibility for bioconversion of the organic matter is increased. Thus in his study, it was observed that ultrasonicated food waste achieved 1.21 times higher methane yield than unsonicated food waste.

#### 4 Conclusions and Recommendations

In this study of work, a combination approach of ultrasonic pretreatment, followed by BMP tests of fruit and vegetable waste was conducted. It is concluded that sonic pretreatment improves the biogas yield of FVW and that the higher sonication time provides a higher methane yield due to an increase in hydrolysed lignocellulose and improved organic matter solubility. This confirms the applicability of this approach.

It is recommended that further studies be performed on a range of sonication times to find the optimum one.

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