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Biogas Development of Tea Dregs and Cow Manure: Study of CN Ratio and Pretreatment on Liquid State Anaerobic Co-Digestion

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ABSTRACT. Energy problems in Indonesia are issues that are not easy to solve. If the energy needs dominated by fossil fuel oil continue to increase without any changes in the pattern of energy use, then Indonesia's sustainability and energy security will be disrupted. Therefore, Indonesia really needs alternative energy. Biogas is an alternative energy produced from the anaerobic degradation of organic compounds and can be a substitute for natural gas and fossil fuels. Tea dregs can be used as a substrate from anaerobic co-digestion biogas production with cow dung. In general, the purpose of this study is to produce biogas from tea pulp and cow manure by anaerobic co-digestion and have a specific purpose, namely (i) Assessing the effect of pretreatment on the yield of biogas produced, (ii) Assessing the effect of pH on yield of biogas, (ii) Assessing the effect of the value of the C/N ratio on the yield of the biogas produced. This research was conducted by making variations in treatment including physical pretreatment with and without grinding (± 1 mm), biological pretreatment with and without addition of 5% v/v microbial consortium, pH controlled (addition of buffer) and uncontrolled, and ratio of C/N tea dregs (25 and 30). The biogas formation process is carried out for 2 months at room temperature with the quantitative response test results in the form of biogas volume every 2 days. Biogas production in pretreatment tea grounds gives better results than without pretreatment. Optimum biogas production is obtained at a C/N ratio 30. Comparison of C/N substrate will affect the growth of microorganisms, the microbes that play an anaerobic process need nutrients to grow and develop, in the form of carbon and nitrogen. The highest biogas accumulation produced was 73.167 ml/gTS on variable C/N ratio 30, NaOH pretreatment, microbial consortium and smooth size of tea dregs that used.

Keywords: biogas, pretreatment, ratio C:N, tea dregs

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1. Introduction

The energy problem in Indonesia is an issue that is not easy to solve. This is of course due to energy consumption which is still dominant in oil and gas. In fact, the national petroleum reserves as of January 1 2015 proved to have decreased by 1.2% compared to the previous year. On the other hand, the rate of fuel consumption as a processed product continues to increase. Meanwhile, the development of production during the last 10 years has shown a downward trend, from 287.30 million barrels or around 800 thousand barrels per day in 2006 and to 251.87 million barrels or 690 thousand barrels per day in 2015. The decline in production was due to wells. Petroleum production is generally old, while production of new wells is still relatively limited (Prasodjo et al., 2016). If the energy demand which is dominated by fuel oil continues to increase without any change in energy use patterns, then Indonesia's energy sustainability and security will be disrupted (Yudiartono et al., 2018). To achieve national energy independence and resilience, as stated in the main regulation, Government Regulation no. 79 of 2014 concerning National Energy Policy, Perpres RUEN outlines the priorities of Indonesia's energy development which includes several things, namely the maximum use of renewable energy by taking into

account the economic level, minimizing the use of petroleum, optimally utilizing natural gas and new energy, and making coal a mainstay national energy supply. Therefore, Indonesia needs to develop new and renewable energy as a solution to these problems.

Biomass energy, which has become a global concern in recent years, is a renewable resource that can be converted into three phases of fuel: gas, liquid and solid (Mao et al., 2018). Indonesia is a country that is rich in both biological and vegetable natural resources. Indonesia, which is said to be one of the countries with the richest biomass potential, still has a fairly low utilization of biomass potential (Biddinika et al., 2016). Most of the biomass energy in Indonesia is also used for household, agriculture, timber and sugar industry, in rural areas for cooking, lighting, rice milling, drying agricultural products, and heat and power generation (Singh and Setiawan, 2013). One of the biomass-based alternative energies that can be applied in Indonesia is biogas.

Biogas is produced from the anaerobic degradation of organic compounds and can be a substitute for natural gas and fossil fuels. Biogas has three main components, namely methane (CH₄), carbon dioxide (CO₂) and nitrogen (N₂). However, there are also very few side products, namely hydrogen sulfide (H₂S), hydrogen (H₂), nitrogen (N₂), ammonia

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(NH_3), oxygen (O_2) and carbon monoxide (CO) (Ullah Khan et al., 2017). Biogas is renewable energy, which means it can be renewed (Manyuchi, Mbohwa and Muzenda, 2018). Turkey produces nearly 2 trillion m^3 per year of biogas and generates 9 million MWh of electricity (Aksay, Ozkaymak and Calhan, 2018). By using cheap waste, even free. One of the processes for making biogas comes from various organic substrates and usually uses an inorganic co-digestion process (López González, Pereda Reyes and Romero Romero, 2017).

Indonesia is the fifth largest tea producing country in the world with a tea production figure in the form of dry leaves of 144.02 thousand tons in 2016 (Zikria, 2017). However, to meet domestic needs, Indonesia still imports tea from several countries (Syaipulloh, 2015). A large number of tea leaves and tea leaf residues are disposed of into the environment through daily tea drinking, instant tea extraction and ready-to-drink tea (Mahaly et al., 2018). The high consumption of tea in Indonesia also causes a large amount of waste produced in the form of tea dregs. Tea leaves contain beneficial amino acids, proteins, vitamins, tannins and polyphenols. After steeping the tea leaves, some nutrients remain in the tea leaves (Aksay, Ozkaymak and Calhan, 2018). So far, the utilization of tea dregs waste is still limited, so it needs development.

Aksay et al. (2018) said that dry tea leaves are brewed with hot water then the result can be said to be waste. The waste generated in large quantities is disposed of with a mixture of other domestic waste which has a high moisture content which can cause environmental pollution and unpleasant odors. Tea dregs waste continues to increase depending on the amount of consumption. Therefore, it is necessary to develop a new method for reusing tea waste. Tea leaves contain amino acids, proteins, vitamins, tannins, polyphenols. After the tea leaves have been brewed, the nutritional content is still left in the tea dregs. Therefore, this waste can be used for biogas production in addition to being used as animal feed and other products.

The population of beef cattle in Indonesia is estimated at 10.8 million head and dairy cattle 350,000 - 400,000 head and if one cow on average every day produces 7 kilograms of dry manure, the dry manure produced in Indonesia is 78.4 million kilograms of dry manure per day (Budiyanto, 2011). One cow per day produces manure ranging from 8 - 10 kg per day or 2.6 - 3.6 tonnes per year (Sholihul and Wikanta, 2017). If this abundant cow dung is not managed properly, this waste will not only affect milk production and quality, but also the surrounding environment (Dianawati and Mulijanti, 2015). Of all ruminant livestock commodities, cattle are the producer of CH_4 which is more than other ruminants (Syarifah and Widiawati, 2017). It has been reported that the highest contributor to CH_4 gas emissions in Indonesia in the livestock sub-sector is beef cattle, which is 65.12% of ruminant livestock emissions, or 58.84% of the total CH_4 gas emissions of all livestock commodities. Cow manure has a high nitrogen content and is due to pre-fermentation in ruminant stomachs, and has been observed to be the most suitable material for high biogas yields through years of research (Verma et al., 2014). Cow manure is an excellent substrate for biogas production by co-digestion with other types of waste materials such as organic industrial waste, household waste and sewage sludge even though the yield of methane as a single substrate is low (Yohaness, 2010). However, cow manure serves as an excellent "carrier" substrate during mixed waste mixing and allows the concentrated inorganic digestion of industrial waste, which would be difficult to handle in isolation. A large number of anaerobic bacteria in cow manure is effective in reducing the organic fraction of livestock manure even though the pH is not

regulated (Shehu, Ibn and Ismail, 2012). Therefore, this research will study the process of making biogas by anaerobic co-digestion from cow dung and tea dregs.

2. Materials and Methods

The research method used to achieve the objectives of this study by conducting laboratory-scale experiments conducted at the Chemical Engineering Waste Management Laboratory, Faculty of Engineering, Diponegoro University. The process and processing method are carried out using the anaerobic digestion method in the mesophilic state, namely at ambient temperature ($25\text{-}35^\circ\text{C}$) by using the feed input with a batch system. The materials used are flag tea dregs collected from Padang Restaurant, cow dung taken from the Faculty of Agriculture and Animal Husbandry, Urea, Microbial Konsortium, NaOH, HCl and Water. The tools used are plastic bottles (biodigester), beaker glasses, stirrers, plastic hoses, water reservoirs, pH indicators, valves, measuring cups, hot glue gun, filters, porcelain cups, measuring flasks.

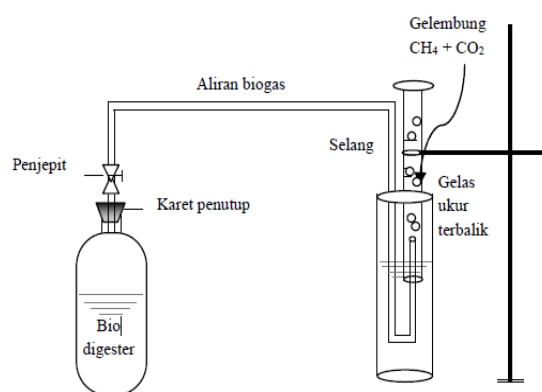


Fig.1 Series of Research Tools

The research design can be seen in Figure 2.

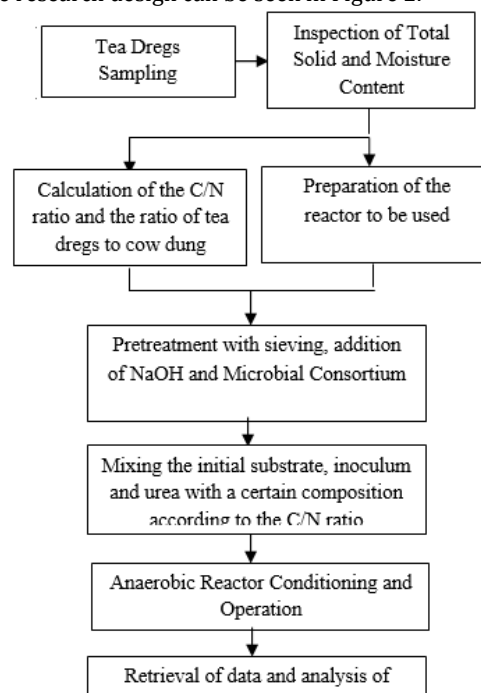


Fig.2 Research Design

Control Variable

1. Operating conditions (batch)
2. The initial pH is neutral (7)
3. Time of observation (for 40 days)
4. Environment temperature (25-35oC) and atmospheric pressure (1 atm)
5. Chemical pretreatment with NaOH 4% g/gTS
6. Reactor volume 600 ml
7. The ratio of the amount of tea dregs and cow dung is 1: 1
8. Liquid state 10% gTS/V

Independent variable

1. Physical pretreatment by sieving ± 1 mm in order to obtain a fine size (through the sieve) and coarse (not through the sieve).
2. Biological pretreatment with the addition of a 5% v/v microbial consortium and without the addition of a microbial consortium.
3. Chemical pretreatment with the addition of NaOH and without the addition of NaOH
4. C/N ratio (25 and 30)

Dependent variable

The response observed from this study was the total biogas volume every 2 days for 40 days.

Preparation phase

The tools are prepared and designed according to the research variables. Take tea dregs and cow dung with a ratio of 1: 1 weighing 60 grams of total solid each and mix it with water to a volume base of 600 ml then put in a container as much as the total variable amount (16 variables).

Data Collection and Analysis Stage

Biogas volume data were analyzed in the form of a graph of the relationship between biogas volume and time. Graphical data analysis and the theory of phenomena that occur on the effect of pretreatment and comparison of the ratio of tea dregs to cow dung respectively on the results obtained in the graph of the results of the study.

3. Results and Discussion

The Effect of the Addition of NaOH to Tea Dregs on Biogas Production

In this research, the biogas produced was viewed from chemical pretreatment with the addition of NaOH. NaOH is mixed with the substrate to accelerate the delignification process or the lignin removal process. In Figure 3 and Figure 4 it can be seen the comparison of the resulting biogas between coarse tea dregs with the addition of NaOH with C/N ratio 25 and 30.

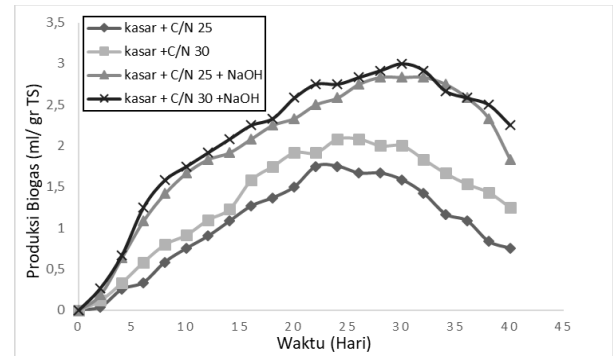


Fig.3 Daily biogas production of crude tea dregs with and without the addition of NaOH at C/N ratios of 25 and 30

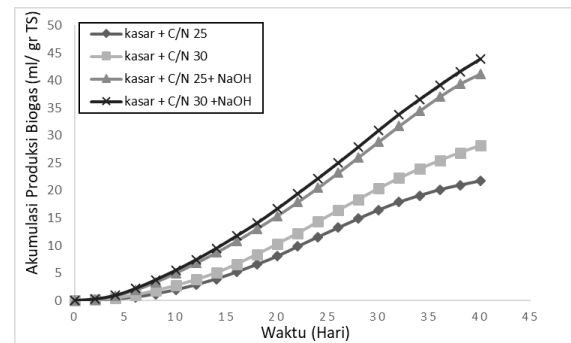


Fig.4 Accumulated production of crude tea dregs biogas with and without the addition of NaOH at C/N ratios 25 and 30

In Figure 3 and Figure 4 we can see the comparison of the amount of biogas production from coarse tea dregs with the addition of NaOH with a difference in the ratio of C/N 25 and 30. The total production of crude tea dregs biogas without addition and with the addition of NaOH at a C/N ratio of 25 respectively were 21.73 ml / g TS and 41.23 ml/gTS, while the total production of crude tea waste biogas without addition and with the addition of NaOH at a C/N ratio of 30 were 28.13 ml/gTS and 43, respectively. 85 ml/gTS.

Giving NaOH which is useful for helping the process of destroying the lignin structure is usually called the delignification process (Kumar et al., 2009). Alkali pretreatment uses bases, with NaOH being one of the most popular, to make the lignocellulose matrix easily degraded by microbes, through removal of the lignin and hemicellulose portions. In addition, it can also reduce the degree of polymerization, crystallinity, and can break the chain between lignin and other polymers (Zheng et al., 2014). The main mechanism of this method is saponification and cleavage of the relationship between lignin and carbohydrates (Mancini et al., 2018). The efficiency of alkaline hydrolysis depends on the substrate and treatment conditions as it is more effective for low lignin biomass such as agricultural residues (Singh et al., 2014). NaOH particles will enter the material and break down the lignin structure so that lignin dissolves more easily which results in a decrease in lignin levels (Elwin, Lutfi, & Hendrawan, 2013).

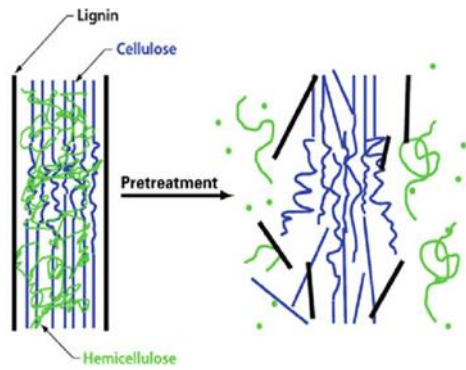


Fig.5 Schematic of the process of destroying the structure of lignin (Xiao et al., 2012)

Lignocellulose consists of 3 main components including cellulose, hemicellulose, and lignin. Among the three main elements, cellulose and hemicellulose are polymers of sugar and can be hydrolyzed. Meanwhile, lignin forms a protective layer that limits the biodegradability of cellulose and hemicellulose. In the delignification process, a certain amount of lignin will be dissolved. This process is a saponification process against the intermolecular ester bonds that surround xylan, hemicellulose and other components, such as lignin and other hemicelluloses. The delignification process causes damage to the lignin structure and releases carbohydrate compounds (Zheng et al, 2009). The process of destroying the structure of materials containing lignocellulose is one of the steps to convert lignocellulose into sugar compounds. The delignification process is believed to be a potential process as a preliminary process in the raw material preparation stage (Taherzadeh and Karimi, 2008). Therefore, the raw material for tea dregs that have been pretreated with NaOH produces more biogas than the others.

NaOH pretreatment can increase the breakdown of cellulose and sugar degradation which is more significant than acid pretreatment, but its application is constrained by a high cost (Talebniya et al., 2010). Pretreatment of NaOH solution against lignocellulosic material causes swelling or swelling, increased deep surface area, reduced degree of polymerization, reduced crystallinity, separation of structural bonds between lignin and carbohydrates and disruption of lignin structure (Fan et al., 1987).

The Effect of Addition of Microbial consortium to Tea Dregs on Biogas Production

This study aims to determine the amount of biogas produced by biological pretreatment, namely by adding a microbial consortium. Pretreatment is carried out to accelerate the degradation process of cellulose. Figure 6 and Figure 7 are the results of biogas between coarse tea dregs with the addition of microbial consortium at a C/N ratio of 25 and 30.

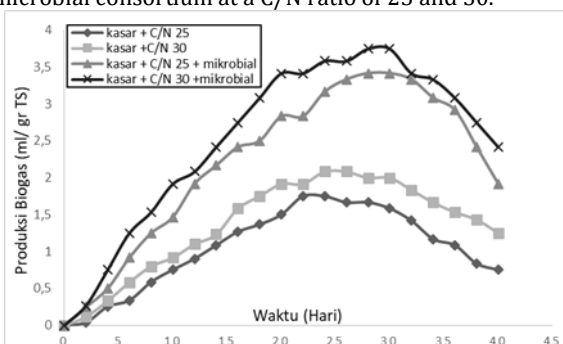


Fig.6 Daily production of crude tea dregs with and without the addition of microbial consortium at C/N ratios of 25 and 30

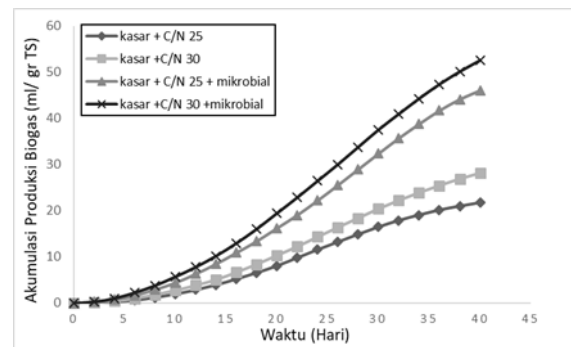


Fig.7 Accumulated production of crude tea dregs biogas with and without the addition of microbial consortium at C/N ratios of 25 and 30

In Figure 6 and Figure 7, it can be seen the comparison of the amount of biogas production from coarse tea dregs at a C/N ratio of 25 and 30 with the addition of a microbial consortium and without a microbial consortium. The total biogas production at C/N 25 ratio with the addition of microbial consortium and without microbial consortium for coarse tea dregs reached 46.05 ml/gTS and 21.73 ml/gTS, respectively. Meanwhile, the total biogas production at C/N ratio of 30 with the addition of microbial consortium and without microbial consortium for coarse tea dregs reached 52.55 ml/gTS and 28.13 ml/gTS, respectively.

The production of biogas in tea dregs with the addition of microbial consortium is better than the production of biogas in tea dregs without the addition of a microbial consortium. This is due to the activity of the microbial consortium which consists of eight main microbes, consisting of the anaerobic genera of Clostridium and Thermoanaerobacterium along with facultative Rhodospirillum rubrum / anaerobic cyclaceae bacteria bacilli, and uncultured bacteria (Tuesorn et al., 2013). Microorganisms also perform delignification functions, reduce the degree of polymerization of cellulose, and hydrolysis of hemicellulose. The addition of microbial consortium accelerates the degradation of cellulose, hemicellulose and lignin into compounds needed by biogas-producing microorganisms, so that biogas production increases (Zhang et al., 2011). Biological pretreatment using a microbial consortium produces a higher yield of biogas with a minimum energy and minimum chemical process compared to other methods (Yuan et al., 2012).

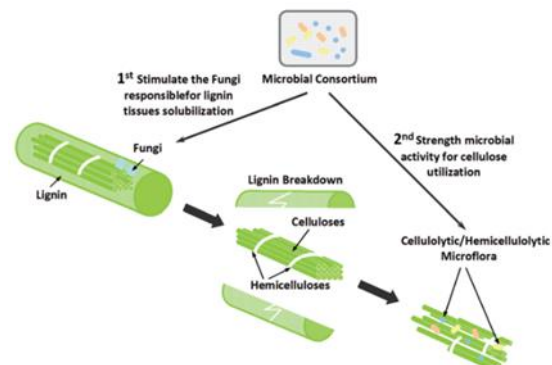


Fig.8 Schematic of the process of destruction of the lignin structure (Zhong et al., 2016)

The general structure of lignocellulose can be explained in Figure 8 where cellulose is wrapped with hemicellulose and

lignin as the main barrier to cellulose, and the destruction of the structure by removing lignin is beneficial for cellulose utilization (Zhong et al., 2016). The strong binding to lignocellulose can be compromised by destroying most of the lignin and the accessibility of cellulose can be improved well after pretreatment using a microbial consortium. In Figure 8, it can be seen that structural destruction with an enlargement of the specific surface area can be achieved through this pretreatment and structural disturbances increase the accessibility of the remaining cellulose (hemicellulose) (Zhong et al., 2016).

Effect of Tea Dregs Size on Tea Dregs on Biogas Production

Pretreatment can increase the digestibility of lignocellulose which is inhibited by several factors, such as: lignin content and composition, cellulose crystallinity, degree of polymerization, pore volume, acetyl groups bound to hemicellulose, surface area and biomass particle size (Alvira et al. 2010). Figure 9 and Figure 10 can be seen the comparison of the biogas production from the waste with a C/N ratio of 25 and 30 ratio of the size of the tea dregs (coarse and fine).

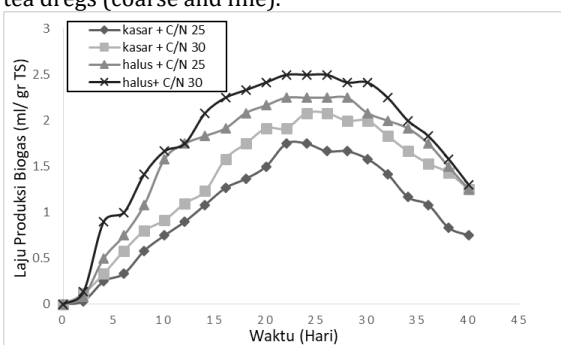


Fig.9 Daily biogas production from tea dregs C/N ratio 25 and 30 with ratio of tea dregs size (coarse and fine)

Figure 9 shows the daily production rate of tea dregs with a C/N ratio of 25 and 30 ratio of tea dregs size (coarse and fine). Biogas production began to occur on the 2nd day for all variables. In crude tea dregs, the C/N ratio 25 of biogas production occurs until the 23rd day, then decreased until the 37th day. In crude tea dregs, the C/N ratio of 30, biogas production occurs until the 22nd day then fluctuates until the 28th day, then decreases until the 40th day. On the fine tea dregs, the C/N ratio 25 of biogas production occurs until the 28th day, then decreases until the 40th day. On the fine tea dregs, the C/N ratio of 30, the biogas production occurs until the 30th day, then it has decreased until the 40th day. The highest biogas production was refined tea pulp with C/N 30.

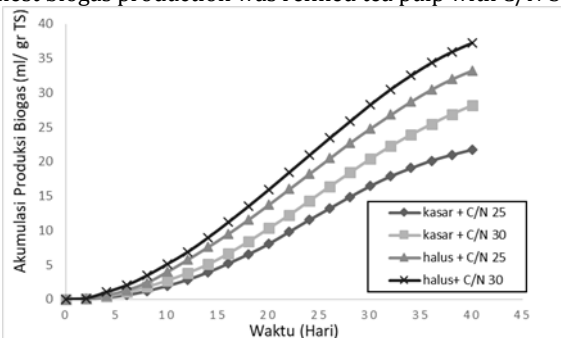


Fig.10 Accumulation of biogas production from tea dregs with C/N ratio 25 and 30 with ratio of tea bag size (coarse and fine)

Figure 10 shows the total biogas production from tea dregs with a C/N ratio of 25 and 30 with a ratio of tea dregs size (coarse and fine). In crude tea dregs, the ratio of C/N 25, the total volume of biogas is 24.97 mL/gTS. Crude tea dregs ratio C/N 30, total volume of biogas 28.2167 mL/gTS. In the tea dregs, the ratio of C/N 25, the total volume of biogas was 34.2167 mL/gTS. In the crude tea dregs, the ratio of C/N 30, the total volume of biogas was 37.383 mL/gTS.

Physical pretreatment is carried out by grinding and making a part of the substrate smaller or pressing it to destroy its cell structure, increasing the specific surface area of the biomass. This pretreatment may better allow enzymatic destruction, especially on lignocellulosic substrates. Particle reduction not only increases the rate of enzymatic digestion, but also reduces the viscosity in the digester (which facilitates stirring) and can reduce the problem of floating layers (Montgomery and Bochmann, 2014).

The sample size can affect the porosity which then affects the contact with the delignifier (Sun & Cheng, 2002). In addition, reducing the sample size will break the long polymer chain into a shorter polymer chain, thereby facilitating the separation of lignin from cellulose bonds (Heradewi, 2007; Permatasari et al., 2014).

This is because the reduction in particle size can reduce the crystallinity of cellulose and disrupt the defense against lignin. Thus, it will facilitate the hydrolysis process resulting in increased bioenergy production (Li et al., 2010). In addition, reducing the size of tea dregs can increase the surface area and pore size, and reduce the crystallinity and degree of polymerization of cellulose (Taherzadeh and Karimi, 2008).

4. Conclusion

Biogas production in pretreated tea dregs was better than without pretreatment. The optimum biogas production is obtained at a C/N ratio 30. The C/N ratio substrate will affect the growth of microorganisms, microbes that play a role in the anaerobic process require nutrients to grow and develop, in the form of a carbon source and a nitrogen source. The highest biogas accumulation produced was 73.167 ml/gTS at the C/N ratio variable 30, NaOH pretreatment, microbial consortium and the size of fine tea pulp. Continuous research needs to be done to determine the length of fermentation for biogas production, test the gas content produced to determine the content of methane gas and other gases, test the fuel value of the biogas produced.

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