

Using Sawdust to Treat Synthetic Municipal Wastewater and Its Consequent Transformation Into Biogas

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ABSTRACT

Sawdust, as an agricultural waste which is highly efficient, readily available, and relatively inexpensive, has the potential to be an applicable alternative adsorbent for the total organic carbon (TOC) removal from synthetic domestic wastewater. This study aims firstly to investigate the feasibility of sawdust as a new adsorbent and understand its adsorption mechanism for TOC. The impact of particle size, pH, contact time, and temperature has been evaluated as the controlling factors on the adsorption process. The results presented that the removal efficiency rose with the decrease of particle size, pH, and temperature, as well as the increase of the contact time. The maximum adsorption was obtained at particle size of 0.05 mm, pH of 1, contact time of 1.5 h, and temperature of 15°C, respectively. The second aim of this study is to utilize the sawdust that is used in the adsorption process as biomass in batch anaerobic digestion (AD) to produce methane. Spent sawdust was characterized by the methane production which was 5.9 times greater than in the case of raw sawdust. Four operating parameters were checked, Carbon/Nitrogen ratio (C/N), inoculation, particle size, and total solid (TS) content. The batch results indicated that the optimum parameters were: 20%, 30%, 2 mm, and 15%, respectively.

Keywords: Anaerobic digestion; sawdust; adsorption; total organic carbon

INTRODUCTION

The three main problems faced by the world today are water, food, and energy supply. In order to solve these problems, the domestic wastewater is now treated as a resource rather than as a waste (Benetti, 2008). The 1st and 3rd problems can be addressed through the use of treated wastewater for domestic consumption, including landscape and crop irrigation, which is widely accepted and used to save water as well as utilize the fertilizing elements it contains. The domestic wastewater can be used as a source for energy through anaerobic digestion, to solve the 3rd problem, which involves the methane gas (CH₄) production from wastewater organic content by anaerobic conversion (Speece, 2007). The organic fraction in wastewater is the most direct and commonly exploited energy source. This fraction is a diverse mixture of molecules with a varied structure and molecular weight. It contains ~42% dissolved organic

carbon, ~27% settleable organic carbon, ~20% supracolloidal organic carbon, and ~11% colloidal organic carbon (Rickert & Hunter, 1971). If these compounds are not removed properly from wastewater through the wastewater treatment plants (WWTPs), and are discharged into water resources (rivers or lakes), they may constitute an environmental problem. This problem has a harmful effect on the water quality and also affects the aquatic life (Huerta-Fontela et al., 2011).

WWTPs remove or minimize the nutrients such as carbon, nitrogen, and phosphorus. Most of the existing treatment processes involve advanced methods (likes activated carbon, photo-oxidation, ozonation, and UV radiation) because the biological treatment alone is not sufficient (Ahn & Logan, 2010; Katsoyiannis & Samara, 2005). Nevertheless, these advanced methods using ozonation and UV light are expensive since they require greater technical experience and are characterized by high energy consumption. Addi-

tionally, when activated carbon is used, the steps of its activation and regeneration are delicate and too costly; moreover, additional tertiary filtration is frequently required (Shukla et al., 2010; Yin et al., 2007). In order to treat wastewater by removing nutrients such as TOC efficiently, new waste materials, especially agricultural wastes like sawdust, wood chips etc., have the economic advantages of obtaining low-cost feedstocks for wastewater treatment and minimizing waste disposal costs. In addition, it offers environmental advantages like reusing and reducing wastes and energy recovery (Yang et al., 2017).

Although sawdust, like any biomass, can be explored as a biogas source, it is far less used as a feedstock for biogas production (Castoldi et al., 2017). Therefore, in this study, it is used for the removal of TOC from wastewater and then for biogas (CH₄) production by anaerobic digestion process. Anaerobic digestion (AD) is the process of converting biomass into biogas energy. This process was used for the production of renewable energy so as to achieve stable energy alternatives that will meet the world demand while mitigating the climate change through reduction of emissions. AD constitutes an economical, eco-friendly, renewable energy source which can produce bio-fertilizers as a by-product (Kimming et al., 2011; Lizasoain et al., 2016).

The main objectives of the present study were to (i) investigate the effects of the adsorption process parameters (particle size, dose, temperature, reaction time, and wastewater pH) on the TOC removal by sawdust and (ii) evaluate its potential biogas production by AD of sawdust after the adsorption process; the influence of different operation factors (particle size, inoculum percent, C/N ratio, and T.S content) was checked as well.

MATERIALS AND METHODS

Raw materials

Our main material (pine sawdust) was brought from a furniture factory of Huazhong University of Science and Technology (HUST), Wuhan, China. The pine sawdust was naturally dried for eight days, then crushed and sieved to four different particle sizes. The ultimate and proximate analyses of sawdust are given in Table 1 (wt%, air-dried basis). Synthetic wastewater in this study was prepared to simulate the characteristics of Chinese municipal wastewater. Its composition was

as follows (mg/L): Glucose (332), NH₄Cl (210), KH₂PO₄ (21.95), MgSO₄ (50), ZnCl₂ (50), and a 1 ml trace element mixture, consisting of 0.075 g CaCl₂, 0.04 g CuCl₂·H₂O, 0.048 g NiCl₂·6H₂O, 0.044 g FeSO₄·7H₂O and 0.120 g H₃BO₃. In order to keep the pH of the synthetic wastewater at 6.8–7.2, 100 mg/L NaHCO₃ was added. The inoculum used for anaerobic digestion process with a total solid content (TS%, 4.25), volatile solid content (VS%, 69.0) and pH (7.2) was collected from the active mesophilic anaerobic reactor in the same lab that treated pig manure to produce biogas. All chemicals and reagents used in this study were of analytical grade.

Batch adsorption process

In order to study removal of TOC from synthetic domestic wastewater by pine sawdust, adsorption experiments in batch mode were carried out. The effects of different factors, including particle size (0.05–1 mm), pH (1–9), adsorbent dose (0.1–0.9 g/50 ml), contact time (10 min – 4 h), and temperature (15–50 °C) were investigated. The pH of the solution was adjusted with 0.1 M HCl and 0.1 M NaOH. The weighted adsorbents were added to 50 mL of wastewater in a 150mL Erlenmeyer flask and then mixed at 200 rpm for the desired contact time at a specific temperature in a water bath shaker. At the end of adsorption process, and after centrifuging solutions, the suspensions were separated; afterwards, the TOC concentrations were measured according to a standard method (APHA). The adsorption capacity and percentage removal of TOC by sawdust were determined by Eqs. (1) and (2), respectively:

$$q_e = \frac{C_o - C_e}{m} \quad (1)$$

$$\% \text{removal TOC} = \frac{C_o - C_e}{C_o} \times 100 \quad (2)$$

where q_e (mg/g) represents the adsorption capacity.

C_o (mg/L) and C_e (mg/L) is the initial and final concentration of TOC, respectively.

V (ml) is the volume of TOC solution,

m (g) is the weight of the dried adsorbents.

Anaerobic digestion process

The anaerobic digestion experiments were conducted in a batch mode using 1 L Duran glass bottles with a working volume of 0.6 L. Two

Table 1. Characteristics of pine sawdust

Ultimate analysis ^a					Proximate analysis ^b			
Carbon	Hydrogen	Nitrogen	Oxygen ^c	Sulfur	Moisture	Volatile	Ash	Fixed carbon
47.15	6.22	0.18	45.20	0.16	6.95	78.01	1.09	13.95

groups of batches were conducted based on the objectives of this study; the first one was fed with raw sawdust and the other was fed with sawdust used in the adsorption process. At the same time, the bottles only fed with the inoculum were used as a control. All the experiments were conducted in duplicate. Four different parameters were studied in these batch tests, i.e. substrates particle size (0.3, 0.8, 1.0, and 2.0 mm), inoculum percent (10, 20, and 30%), carbon to nitrogen (C/N) ratio (20, 25, and 30%), and total solid (TS) content (5, 10, and 15%). Each parameter was studied alone and others were kept fixed. After adding the designed amounts of sawdust and inoculum in the batch digesters, the C/N ratio and TS% were checked. Prior to sealing, each bottle was flushed with nitrogen gas (99.9% purity) for 2–5 min to confirm anaerobic conditions. Then, the bottles were tightly closed with rubber stopper and caps and placed in a water bath shaker at (35°C) and stirred at 100 rpm. During the batch test, the daily biogas produced in the reactor flowed through a bottle filled with 3M NaOH solution for CO₂-fixation and remaining methane (CH₄), which was measured with the water displacement method.

Analytical methods

The water quality parameters, including total organic carbon (TOC), total Kjeldahl nitrogen (TKN), total solids (TS) and volatile solids (VS) were determined according to Standard Methods (Federation & Association, 2005). The pH of the solution was measured using a PHS-25C pH meter made by Shanghai Precision & Scientific Instrument Co., Ltd. The carbon and nitrogen analysis was conducted using a Vario EL (element analyzer) made by Elementar Analysensysteme GmbH. The CH₄ content was analyzed via gas chromatography (SP-2100, China).

RESULTS AND DISCUSSIONS

Adsorption results

Adsorptive removal of TOC was studied for four different particle sizes of pine sawdust

equalling $r < 0.05$, $0.2 < r < 0.3$, $0.3 < r < 0.5$, and $0.5 < r < 1$ mm. As shown in Figure 1a, the rate of adsorption was influenced by the size distribution. It is interesting to note that an increase in the TOC adsorption percentage occurs from 13.0% to 46.5% when the adsorbent size decreases from $0.5 < r < 1$ mm to $r < 0.05$ mm. This behaviour can be explained due to the accessibility to larger surface area of the adsorbent for adsorption (Gupta et al., 2015).

In order to study the effect of adsorbent dose in solution on the TOC adsorption percentage, different dosage was used (ranging from 0.1 to 0.9 g/50 mL) and the results are shown in Figure 1b. It was observed that the removal efficiency increases along with the amount of adsorbent in the solution. This can be attributed to the increase in the accessibility of adsorption sites for the TOC. Many studies are in agreement with the current work results (Kazemi et al., 2016; Khamparia & Jaspal, 2016; Mor et al., 2016) where they reported that the sorption of pollutant increases as dosages increased. On the other hand, a reverse trend was observed with the adsorption capacity which decreased as the adsorbent dosage was increased (Figure 1b). The reason of this was possibly due to the particle interaction, such as aggregation, resulting from high adsorbent dosage, which lead to a decrease in the total surface area of the adsorbent and increase in the diffusional path length (Ghaedi et al., 2012).

The pH value of the initial wastewater is the most important parameter affecting the adsorption of TOC. The uptake of TOC by pine sawdust was studied as a function of pH ranging from 1 to 9. The results show that the removal percentage of TOC from the synthetic wastewater decreased with the rise in pH of the solution (Figure 1c). Maximum 62% TOC adsorption was obtained at pH 1 indicating that the TOC removal is more efficient under acidic conditions and it minimizes to 12% with a rise in pH to 9. These results were consistent with the previous studies, where it also showed that the adsorption percentage increased with pH reduction (Khamparia & Jaspal, 2016; Liu et al., 2016; Mor et al., 2016).

The extent of TOC taken up from synthetic municipal wastewater by pine sawdust was addi-

tionally verified as a function of contact time. The contact time was changed from 10 min to 4 h. The results of the contact time effects are shown in Figure 1d. It is clear from Figure 1d that equilibrium was reached after 1.5 h of contact time (removal percent was 36%). Afterwards, as shown in Figure 1d, the TOC removal was decreased to 32%. On the basis of the findings from contact time experiments, 1.5 h was taken as equilibrium time in next adsorption studies.

The impact of temperature on the TOC adsorption by pine sawdust was checked (Figure 1e). Five temperatures were used in this part of adsorp-

tion experiment (15–50°C). As can be seen, the TOC removal percent decreases sharply with the rising temperature from 15°C to 50°C. Maximum TOC removal of 37% was observed at 15°C and decreased to 17% at 50°C. Many studies reported increases in the adsorption percentage along with temperature (El-Naas et al., 2010; Kazemi et al., 2016). This indicates the exothermic nature of the adsorption process. Moreover, the decrease of adsorption with increasing temperature can be explained by the weakening of the sorptive forces between the active sites on the sorbent and the adsorptive species (Mor et al., 2016).

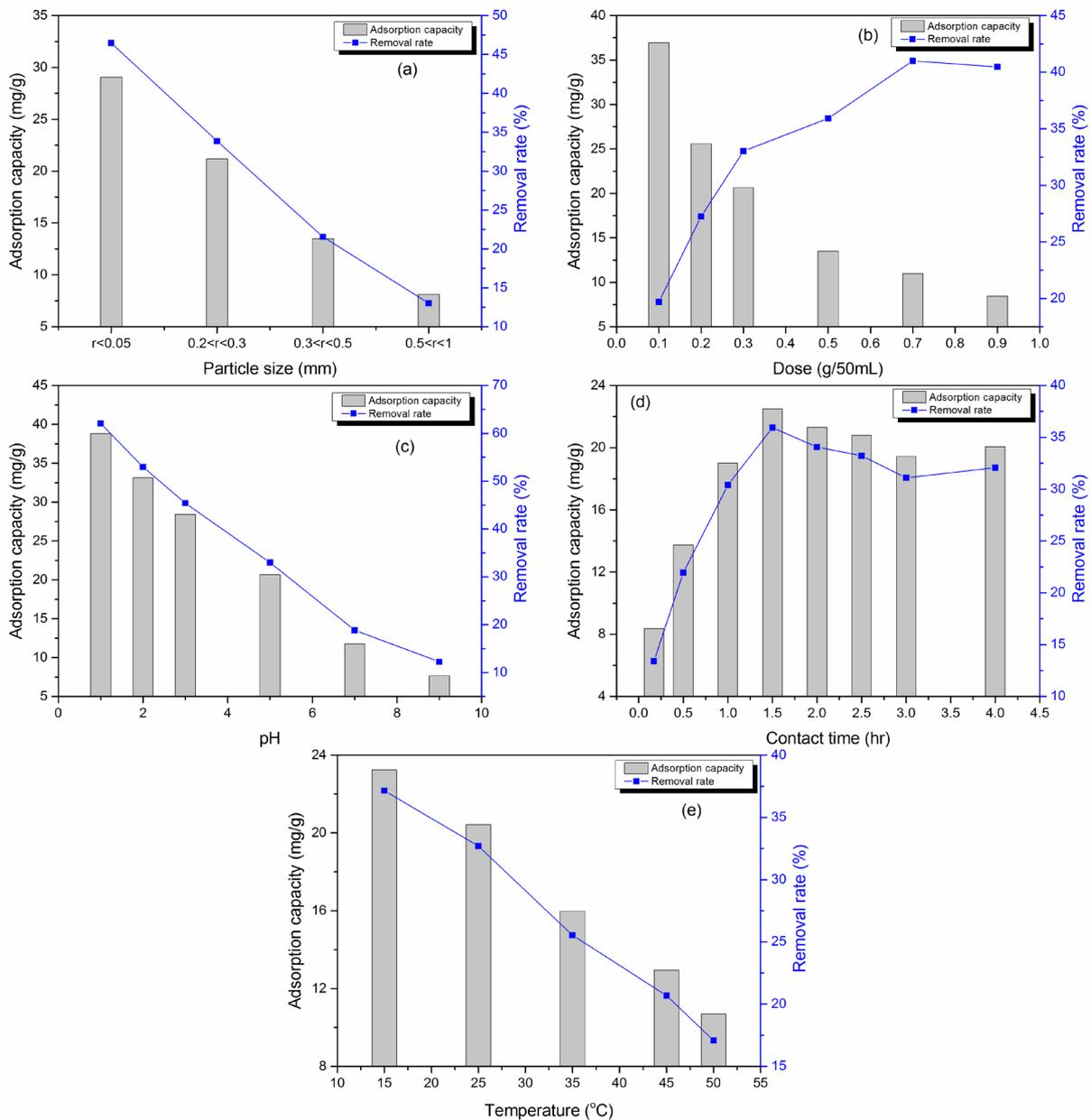


Figure 1. Effect of (a) particle size, (b) adsorbent dosage, (c) pH, (d) contact time, and (e) temperature on adsorption process

Anaerobic digestion results

The second part of this paper was to study the anaerobic digestion of sawdust after using it in the adsorption process. This part was divided into five groups: the first one checked the effect of TOC adsorbed on sawdust on AD of sawdust, while the other four groups investigated the effects of C/N ratio, inoculum percentage, particle size, and TS content on the AD process and biogas production.

In order to study the feasibility of anaerobic digestion of sawdust before and after the adsorption process, batch tests were conducted on both sawdust (raw and spent) and the results were illustrated in Figure 2. It is clear from Figure 2 that the AD of spent sawdust was a very important step of adsorption process because of the great amount of energy that can be recovered from it instead of being wasted. Figure 2 shows that the digestion process of raw sawdust did not take more than 25 days and stopped, while for spent sawdust, the process continued for 45 days. Maximum daily CH_4 produced by spent sawdust was 1250 mL/d, which was 290% more than maximum CH_4 produced from raw sawdust (Figure 2a), this may be due to the large amount of TOC adsorbed on sawdust, which can be digested easily by microorganisms. Figure 2b showed the cumulative CH_4 produced from digestion process. It can be seen that the reused sawdust had a significantly higher CH_4 production (5.9 times) compared to raw sawdust. Therefore, using low-cost biomass has a great theoretical and practical value for treatment of municipal sewage, removal of the hydrocarbon resources from sewage, and finally using absorbent as raw materials to produce biogas energy.

Three different C/N ratios (20%, 25%, and 30%) were carried out to find the optimum ratio. The results were shown in Figure 3. As can be seen in Figure 3, the best C/N ratio was 20% for CH_4 production volume. The batch reactor with C/N of 20% had the maximum daily CH_4 production (1075 mL/d), which was 28% and 55% more than the production of reactors with C/N of 25% and 30%, respectively (Figure 3a). Additionally, cumulative CH_4 produced from a batch with C/N ratio of 20% (8775 mL) was higher than other batches by 14% and 23%, respectively (Figure 3b). The result of this parameter was consistent with the previous studies, where the optimum C/N ratio was 20–30%. In a study conducted by Yen and Brune to investigate the effect of C/N ratio on the anaerobic co-digestion of algal sludge and waste paper, the results showed that the optimized C/N ratio for the co-digestion was 20/1 to 25/1 (Yen & Brune, 2007). Haider et al. (2015) carried out another study to investigate the anaerobic co-digestion of food waste and rice husk. Their results showed that highest specific biogas yield of 584 L/kg VS was obtained from feedstock with C/N ratio of 20 (Haider et al., 2015).

In order to study the effect of inoculum amount on the anaerobic digestion of sawdust, batch experiments with three inoculum amounts (10, 20, and 30%) were conducted. The experiments were continued for 40 days. Figure 4 shows the daily and cumulative CH_4 production of sawdust digestion. As can be seen, the CH_4 production increased along with the inoculum percentage. This can be explained by the fact that when the inoculum amount decreases, it may cause overload, the unfavourable situation where there was too much biomass for the microorganisms to digest.

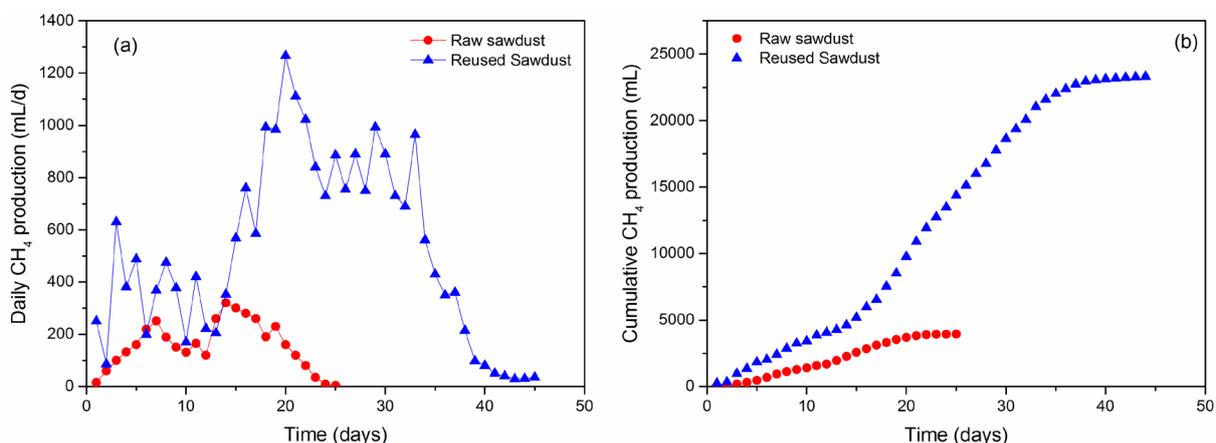


Figure 2. Effect of TOC on AD of sawdust

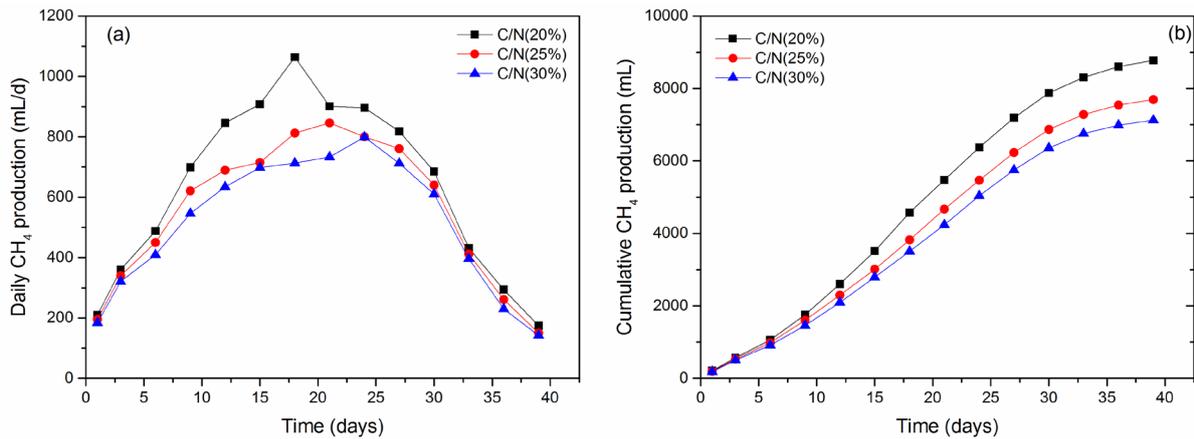


Figure 3. Effect of C/N ratio on AD of sawdust

Thus, higher inoculum could supply some kind of safety factor in the BMP assay since the appropriate inoculum is able to process a higher flow of metabolites (Dechrugsa et al., 2013). Maximum daily CH₄ produced was 1370 mL/d achieved by 30% inoculum amount, which was 52% and 26% more than other inoculum percentages, respectively (Figure 4a). The cumulative CH₄ production after 40 days of AD of sawdust with 10%, 20%, and 30% inoculum percent were 7940, 8275, and 8700 mL, respectively (Figure 4b). This result was comparable to previous studies, where the biogas production increased along with the inoculum amount (Dechrugsa et al., 2013; Haider et al., 2015).

One of the parameters investigated in this study was biomass particle size and its effect on AD of sawdust and biogas produced. As can be seen from Figure 5, the best particle size was 2 mm. Maximum daily CH₄ production (1700 mL/d) was achieved by 2 mm particle size with no significant difference with a 1 mm par-

ticle size (1590 mL/d), but it has a significant difference with other sizes 0.3 and 0.8 mm, 107% and 31%, respectively (Figure 5a). Additionally, the cumulative CH₄ produced during the entire digestion time, the batch with 2 mm particle size had the highest amount of CH₄ generated. This particle size showed a 2.3- and 1.5-fold higher CH₄ volume than 0.3 and 0.8 mm particle sizes, respectively. A small difference (2.5%) was achieved with 1mm particle sizes. These results were in agreement with previous studies (Agyeman & Tao, 2014; De la Rubia et al., 2011; Zhang & Banks, 2013). Zhang and Banks conducted a study to check the impact of different particle sizes on AD of the organic fraction of municipal solid waste. Their results indicated that the digester with the 2 mm mean particle size was characterized by slightly higher methane production than the 4 mm mean particle size (Zhang & Banks, 2013). In another study, Agyeman and Tao found in their work that the maximum methane production rate and specific methane yield were signifi-

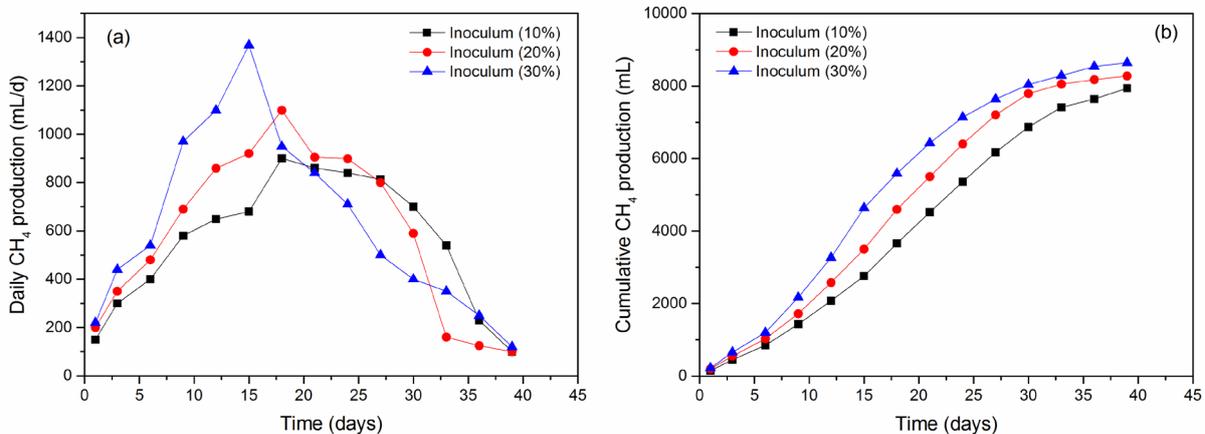


Figure 4. Effect of inoculum percentage on AD of sawdust

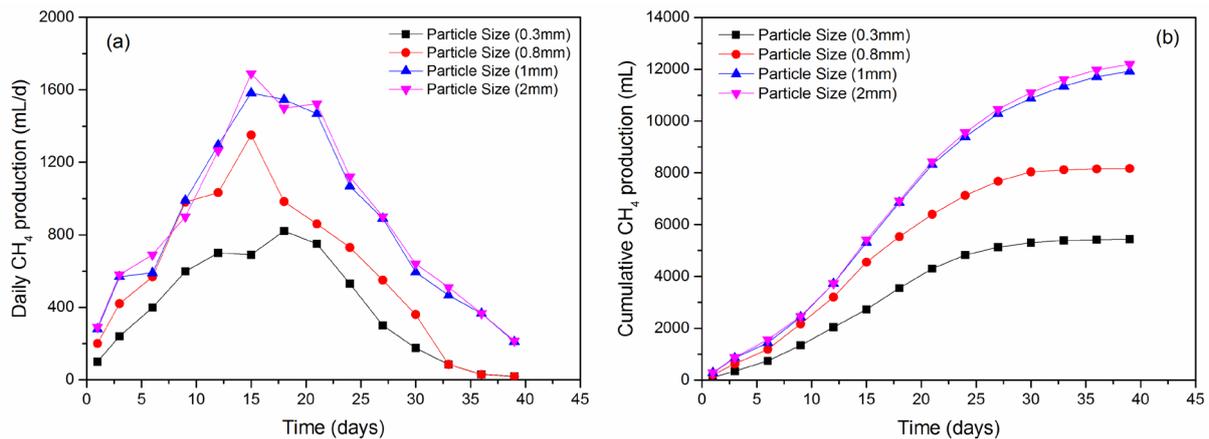


Figure 5. Effect of particle size on AD of sawdust

cantly higher in the digester with 2.5mm (fine) particle size compared to other reactors (Agye-man & Tao, 2014). On the other hand, De la Rubia et al. evaluated three particles size ranges of (1) 0.355–0.55 mm, (2) 0.710–1.0 mm and (3) 1.4–2.0 mm. They showed that the highest methane yield was obtained for the largest particle size analysed (3) (De la Rubia et al., 2011).

The last parameter investigated was total solid (TS) content. Three different contents were checked (5%, 10%, and 15%) and the results were illustrated in Figure 6. As can be seen in Figure 6, the CH₄ production increased along with the TS content. This result was consistent with a previous work (Yi et al., 2014). For daily methane produced, highest methane was achieved by TS content with 15%, which was 1.30 and 1.19-fold higher than other TS contents, 5% and 10%, respectively (Figure 6a). In addition, the cumulative CH₄ production had the same trend with respect to 15% TS content, where 15% TS showed a 28% and 18% more cumulative CH₄ than other

TS contents, respectively (Figure 6b). These results agreed with previous studies, which proposed that the increase of feeding TS contents lower than 20% has a favourable effect on the CH₄ production (Dai et al., 2013; Yi et al., 2014).

CONCLUSIONS

Anaerobic digestion of pine sawdust after adsorption of TOC from synthetic domestic wastewater can increase the recovery of energy products from sawdust. The sawdust used in this study showed a good performance in the adsorption process. The removal efficiency of TOC was increased with the decrease in adsorbent particle size, pH, and temperature. In contrast, the efficiency increased along with the contact time. The main AD conclusions obtained from this study are: the big amount of energy recovered and methane produced from AD of used sawdust demonstrate the importance of this step after adsorption pro-

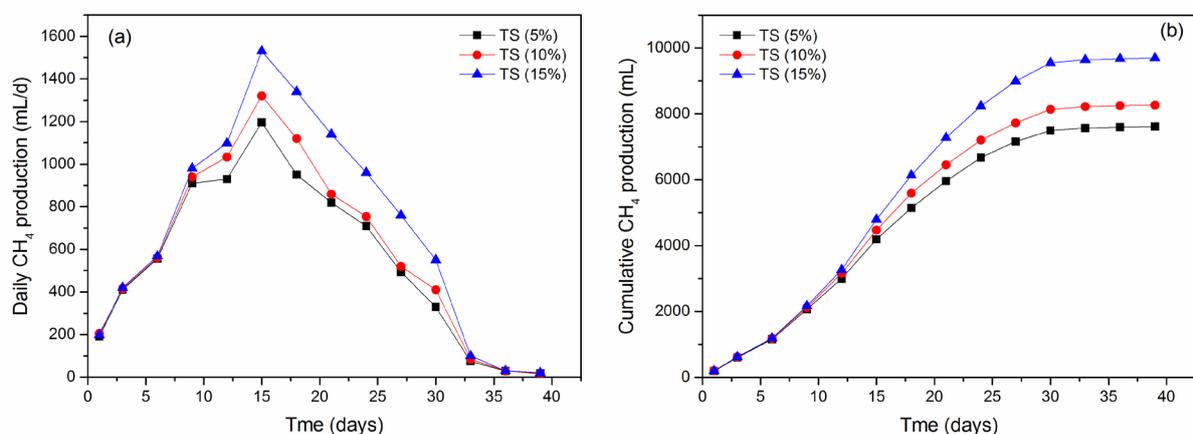


Figure 6. Effect of TS content on AD of sawdust

cess. In addition, the best operating parameters of AD (C/N, inoculation, particle size, and TS) were: 20%, 30%, 2 mm, and 15%, respectively.

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