Low-Temperature Pretreatment of Biomass for Enhancing Biogas Production

Subjects: Agricultural Engineering

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Low-temperature pretreatment (LTPT, Temp. < 100 °C or 140 °C) has the advantages of low input, simplicity, and energy saving, which makes engineering easy to use for improving biogas production. However, compared with high-temperature pretreatment (>150 °C) that can destroy recalcitrant polymerized matter in biomass, the action mechanism of heat treatment of biomass is unclear. Improving LTPT on biogas yield is often influenced by feedstock type, treatment temperature, exposure time, and fermentation conditions. Such as, even when belonging to the same algal biomass, the response to LTPT varies between species. Therefore, forming a unified method for LTPT to be applied in practice is difficult.

Keywords: organic waste treatment ; anaerobic digestion ; thermal pretreatment ; biogas ; heat pretreatment

1. Introduction

People's production and living produce organic waste continually, which has hot spots such as large production, complex composition, and easy-to-breed germs, posing a potential environmental threat [1][2]. Anaerobic digestion (AD) and composting are two main technical methods to treat organic waste [3]. Because it can directly convert high water content materials and recover energy [4] and fertilizer nutrients [5], anaerobic digestion receives excellent attention and has been widely studied and applied over the world [6]. To sum up, the anaerobic transformation of organic waste involves environmental protection, alternative energy production, and emission reduction of total greenhouse gases (GHG) by replacing some fossil energy fuels [7]. However, some organic wastes contain much refractory organic matter (lignocellulosic components) and various microorganisms, so it is challenging to earn ideal or economic biogas production efficiency in an actual trial [8]. Moreover, the investment in biogas projects is high and hard to be compressed because of the need for giant digestion tanks [9] and waste air treatment techniques to avoid emissions and improve biogas quality before use [10]. Therefore, many types of research have been focusing on improving biomass material's conversion efficiency for lower-cost biogas production [11].

Many studies have shown the hydrolysis of organic macromolecules is the key to improving anaerobic biogas production efficiency [12]. Some pretreatment methods have been developed for promoting hydrolysis, such as mechanical, chemical, and thermal [13][14]. The reported specific methods include heat [15][16], ultrasound [17], advanced oxidation [18], alkaline cracking, dry milling, hot water, steam explosion [19], degrease [20], etc. Since the energy input required for heat treatment (HP) can be supplied by choosing thermal energy with a lower energy grade (low ratio of exergy/energy), recovering waste heat energy or directly burning some fuel nearby can be used to provide energy for the HP process. This makes HP cheaper and easy to be applied to practical projects. HP has been successfully applied at an industrial scale and is one of the earliest methods recognized as having the potential to improve AD [21][22] and has been widely concerned and studied in enhancing the bioavailability of biomass. This pretreatment strategy can break up cell membranes resulting in soluble organic substrates that are easily be hydrolyzed during digestion [16][17][18][19]. Moreover, HP can also effectively weaken pathogens' reproduction and decrease feed liquid viscosity [23].

HP is usually performed over a wide temperature range of 50–250 °C and can be divided into two categories according to temperature: high-temperature pretreatment (HTPT) and low-temperature pretreatment (LTPT) [21][24]. The required temperature of HTPT is above 140 °C, and its main purpose is to promote the dissolution or partial dissolution of recalcitrant components and improve the bioavailability of biomass materials [25][26][27]. Such as, previous studies proved that hemicellulose and cellulose solubilize at temperatures >150 °C and 200 °C, respectively [28][29]. While the LTPT employs temperatures below 140 °C (or below 100 °C) for AD improvement [21][30][31][32]. Protot et al. [33] stated that thermal pretreatment below 100 °C can impel the deflocculation of macromolecules. Neyens and Bayens [34] reported that LTPT resulted in the solubilization of proteins and particulate carbohydrates in sludge. Some authors proposed that pretreatment below 100 °C can be considered a biological process since biomass solubilization occurs because of a higher activity of thermophilic and hyperthermophilic bacteria populations [35][36]. In this case, exposure time also plays a more important role [37].

2. General Characteristics of Heat-Treated Biomass

Hydrothermal pretreatment will change biomass material characteristics, such as pH, conductivity, nutrient release, organic matter dissolution, etc. Hren et al. [38] reported pH values decreased during the pretreatments of riverbank grass, sewage sludge, and rumen fluid. The reason may be that thermal pretreatment forms amino acids and fatty acids. Thermal treatment can promote the release of intracellular ions to the outside, which may be the main reason for the change of conductivity, especially in biomass such as grass [38], vegetables [39], microalgae [40], and sludge. Due to the accumulation of nutrients in the process of sludge formation, thermal pretreatment of sludge can effectively release nitrogen (NH_4^+-N) and soluble phosphorus (PO_4^{3-}-P) [32]. Yan et al. [41] reported the concentration of NH_4^+-N increased from 21.0 ± 0.6 mg·L^{-1} to 200.9 ± 2.9 mg·L^{-1} after pretreatment at 100 °C. Many studies have pointed out that increasing the organic matter dissolution is the main reason for pretreatment to improve the hydrolysis step in anaerobic degradation [42]. Rodriguez-Verde et al. [43] found the conducted thermal pretreatment of manure resulted in an increase in the solubilized fraction from 0.20 to 0.38 and 0.43 at 70 °C and 90 °C, respectively. Passos et al. [36] also proved that the pretreatment at 95 °C for ten hours increased VS solubilization of microalgal biomass by 1188%.

3. Municipal Solid Waste (MSW)

With the acceleration and completion of urbanization in developing countries, the amount of MSW is also rising rapidly. Statistics show that more than 4 billion tonnes of solid waste, nearly half of which is MSW, is produced worldwide each year [44]. These wastes have the characteristics of large yield, high water content, and complex composition and are potentially harmful. MSW typically consists of 46% organic content (food/kitchen waste, activated sludge, yard waste, wood, and craft residues), followed by 17% paper, 10% plastic, 5% glass, 4% metal, and 18% others [45]. Many reports have pointed out that the organic fraction of MSW (OFMSW) is an excellent raw material for biogas production because it is rich in organic substances such as starch, protein, and oil [46][47][48]. Thermal pretreatment can also solubilize food waste to improve the AD performance of OFMSW [49]. However, due to the different living habits, the components of OFMSW produced from other regions are various, and improving biogas production by thermal pretreatment will also vary. Thus, it is necessary to summarize the LTPT results of OFMSW to explore the empirical law more suitable for practical applications. Two typical municipal organic wastes, food/kitchen waste and waste-activated sludge (WAS), are used here as the focus of the discussion.

3.1. Food/Kitchen Waste

Due to the high water content and biodegradability, food waste (FW) and kitchen waste (KW) are suitable for AD to produce biogas. In addition, the content of lignocellulosic compounds is low in this biomass, so gentle pretreatment can satisfy the need to improve AD performance. Ma et al. [50] reported an 11% improvement in methane production when pretreating KW at 120 °C for 30 min. Li et al. [51] displayed that temperature coupled with exposure time affects the subsequent improvement of methane production of KW. Such as, biogas production from pretreatment at 70 °C for 90 min is higher than those for a shorter duration (10–60 min), while the treatments at 90 °C and 120 °C obtained the maximum biogas yields lasting 10 and 30 min, respectively. They also pointed out the Maillard reaction would be induced when the pretreatment temperature went up to 140 °C, which reduced the methane yield. This suggests that high-temperature pretreatment is unsuitable for FW and KW, and its high sugar and protein content will lead to the formation of some adverse reactants [52]. Kuo and Cheng [53] conducted thermal treatment of KW at different temperatures to improve hydrolysis and chemical oxygen demand (COD) removal, which showed that pre-treatment at 60 °C yielded the highest total COD (TCOD) removal efficiency (79.2%) after 300 h reaction. The pretreatment at 70 °C obtained the maximum biogas yield of 822 mL·g^{-1} VS in an investigation from 55 °C to 160 °C [51]. Sometimes, thermal treatment hardly improves the cumulative biogas yield, but it can change the biogas production rate. Wang et al. [54] pretreated FW at 70 °C for two hours, and the methane production increased only by 2.7%. Still, the pretreatment halved the time to produce the same quantity of methane compared to the anaerobic digestion of fresh FW. This suggests that heat treatment improves the kinetic features of AD.

Other studies have shown that thermal treatment coupled with chemical reagents can better solubilize KW. Seyed Abbas et al. [55] conducted a thermo-chemical pretreatment on kitchen waste (cooked rice, pasta, ground beef, apple, etc.). They showed that the pretreatment at 120 °C with NaOH 5N can provide the best conditions to increase biogas and methane production. Ma et al. [50] proved that thermal-acid pretreatment at room temperature (pH = 2) obtained a better solubilization rate of kitchen waste than other pretreatments with more severe conditions.

3.2. WAS

The heat treatment of WAS was shown as early as 1970 as an effective pretreatment method for AD [56]. Then many studies have proven that thermal pretreatment can accelerate hydrolysis, shorten sludge's digestion time, and increase biogas production. It has been commercially operational at full scale since 1995 [57]. The temperature range of sludge heat treatment reported in the previous literature is also relatively wide, at 60–270 °C [30]. However, research on LTPT of sludge waste has not been well summarized, and its improvement in biogas production can be more than five times [58]. Appels et al. [37] showed that thermal pretreatment could effectively dissolve both organic and inorganic matter, and the subsequent anaerobic digestion efficiency of sludge at 90 °C, 60 min of pretreatment can be improved 11-fold. Kim et al. [59] found that after 30 min of heat treatment at 121 °C, the damage rate of volatile solids (VS) increased by 30%, and gas production increased by 32%. Nges et al. [60] conducted anaerobic digestion of biogas sludge through experiments and pretreated it at 50 °C for 48 h, resulting in an 11% increase in methane production. For high solid sludge, low temperature thermal pretreatment is also effective, Liao et al. [61] pretreated high solid sludge (TS = 15%) at low temperature (60–80 °C) and carried out intermittent anaerobic digestion experiment and continuous anaerobic digestion experiment and found that low-temperature pretreatment could accelerate digestion of high solid sludge and improve biogas production.

Similarly, applying heat treatment coupled with chemical reagents in sludge pretreatment has also received attention. Xiao et al. [62] conducted high-temperature thermal pretreatment (160 °C) and LTPT by adding alkali (60 °C, pH 12.0) for sludge, respectively, and obtained similar methane production and organic matter removals. This suggests that chemical assistance can compensate for the shortage of pretreatment at a lower temperature. Zheng et al. [63] also reached a similar conclusion through experiments, low-temperature thermos-alkali pretreatment (60 °C, pH 12.0) has better energy efficiency. However, the auxiliary chemical reagent will increase the treatment cost. Appropriate pretreatment only for the substrates with poorer biodegradability before mixed AD could reduce the capital and operating costs [64]. Some studies have also proven that adding chemical reagents in thermal pretreatment may produce some adverse effects. Gunerhan et al. [65] reported that increasing the concentration of NaOH and HCl in thermal pretreatment at 60–100 °C can enhance the COD solubilization of fruit and vegetable harvesting wastes. In contrast, it reduced the concentration of soluble sugar which can be directly converted to methane. It can be concluded from these precious studies that whether heat treatment alone or chemically assisted, the treatment effect depends on the specific substrate characteristics and the set operating conditions.

4. Animal Manure Biomass

Meat, egg, and milk have become essential food for human life worldwide. It is crucial for human nutrition intake and improving living standards: about 270 million dairy cows and 677 million pigs worldwide [66]. Similarly, the annual amount of fecal production is also significant, which has a tremendous potential threat to the human living environment. Recycling energy and fertilizer through AD is helpful for animal manure treatment [67]. However, those initial characteristics of high recalcitrant fibers content, high viscosity, and rich in pathogens are unfavorable to the AD of manure biomass for biogas production. Many facts have proved that the thermal pretreatment method can weaken these adverse factors [12][14][68]. However, due to the differences in chemical composition and physical properties, the reaction results of thermal treatment of different types of animal manures may also be different. For instance, the methane yield of pig manure and sewage waste increased after heating treatment, while that of dairy manure decreased by 6.9% [69]. Therefore, a research of LTPT of manure biomass is carried out according to different categories and mainly focuses on swine/pig manure, cow/dairy/cattle manure, and chicken/poultry manure.

4.1. Pig/Swine Manure

Many studies have shown that thermal pretreatment can significantly improve the biogas production performance of swine/pig manure. Menardo et al. [70] pretreated dehydrated pig manure (PM), digested it at 120 °C and found that methane production increased by 35–171%. Increasing soluble COD may be the main reason for improving biogas production of PM after LTPT. Huang et al. [71] pretreated swine manure (SM) at 110–130 °C for 30 min and achieved a CH_4 yield of 280.18–328.93 mL·g^{-1} VS$_{fed}$ increasing 14–34%. The reason may be the increase of 13–26% in soluble organic carbon concentration after pretreatment. Bonmatí et al. [72] found that the concentration of soluble compounds in pig slurry rose after hydrothermal pretreatment below 90 °C, increasing methane yield. Some studies showed that the inhibitor concentration of PM liquid is low after LTPT, which did not affect biogas production. Fang et al. [73] reported that in both sludge and SM samples, the total biogas and methane productions were enhanced by the 125 °C heating treatment but inhibited by the 225 °C treatment, and they also pointed out the pretreatments at higher temperature may produce inhibitors (e.g., melanoidin). Another study also found the treatment at 100 °C obtained the maximum biogas yield of 0.48 ± 0.02 $L \cdot g^{-1}$ VS, a 30% increase from the raw manure sample. In comparison, biogas production from thermally treated at 130 °C and 150 °C showed less biogas production. The speculated potential reason may be that high temperatures formed complex organic compounds which are difficult to degrade [74]. LTPT can also improve microbial distribution in the AD system of PM. Mladenovska et al. [75] pretreated the mixture of cattle and swine manure at 100–140 °C and obtained an enhancement of specific methane yield in the range of 9–24% and 10–17% for the 20- and 40-min treatment, respectively. Moreover, they also found that continuous feeding of heat-treated PM can affect microbial species richness in a continuous stirring tank reactor and give it the ability to preserve high biogas production.

However, a few studies have also found that LTPT can improve COD’s solubility. Still, biogas production is not significant, which may be due to the high fiber content in the manure samples. Raju et al. [68] found that PM improved biogas production at pretreatment temperatures of 125 °C, while pretreatment at 100 °C did not improve. They also revealed that LTPT has little effect on the cellulose and hemicellulose fractions. Carrère et al. proved that pretreatment of 70–90 °C can only increase the soluble substances and biogas production of the liquid part of PM, while improving the overall biogas production needs a higher temperature of >150 °C [76].

4.2. Cattle/Dairy Manure

The high content of indigestible fiber is an essential feature that distinguishes cattle/dairy manure from other animal manures, while LTPT is hard to decompose these resistant components. As a result, the effective pretreatment temperature of cattle/dairy manure is higher than other manure. Its pretreating temperatures range from 100 to 140 °C, and the exposure time may be required to be extended moderately. Passos et al. [77] found the only conditions that reached methane yield increments were those with long exposure times (i.e., 37 °C for 12 and 24 h), which were 3.6% and 20.5% higher than untreated dairy manure (DM), respectively. At the same time, the thermal pretreatments for 5 and 30 min did not enhance the final methane yield. Wilton et al. [22] reported that the methane production of DM increased by 37% after thermal treatment at 125 °C and for 30 min, while the treatments at a temperature below 125 °C and duration time less than 35 min showed no significant difference in methane production. The reason may be that the bovine gut had previously digested DM, a process that already alters the substrate’s lignocellulosic content, which may render thermal pretreatment redundant.

Therefore, many researchers have been exploring the heat treatment of DM at a higher temperature, mixed with other materials, or assisted by chemical reagents [68][78][79]. Şenol et al. [78] conducted a co-digestion of CM, corn silage, and sugar beet pulp (2:1:1), and pretreating at 100 °C gained a 40% increase in biogas production. Additionally, several studies reported a significant hydrolysis enhancement after heat treatment of dairy/cattle manure by adding chemical reagents (e.g., oxalic acid, sulfuric acid) [80][81][82].

Similarly, inhibition resulting from heat treatment also appears in the AD of dairy/cattle manure, which may occur in both low- and high-temperature cases. Raju et al. [68] found the methane potential of CM decreased by about 10% at the pretreatment condition of 100 °C. Chan et al. [83] reported preheating dairy manure at a temperature below 100 °C and with acid would decrease methane production. Budde et al. [79] found that the abundance of inhibitors and other non-digestible substances led to lower methane yields in the pretreatment at 220 °C than those obtained from untreated CM. In conclusion, higher temperatures, longer exposure time, or chemical reagent assistance may favor the thermal treatment of dairy/cattle manure. Moreover, paying attention to the generation of inhibitors is necessary.

4.3. Chicken/Poultry Manure

Chicken/poultry manure has also been proven to produce biogas. Its yield can reach more than 400 $mL \cdot g^{-1}$ TS [43][84], which is better than pig manure and cow manure, probably because chicken manure (CHM) contains undigested feed. However, the content of organic nitrogen (protein, urea, uric acid) in chicken/poultry manure is higher than that in other manures, mainly due to the need for the rapid synthesis of eggs, feathers, and meat protein. During AD, the organic nitrogen is transformed into ammonia, inhibiting the anaerobic biogas generation process [85][86]. Therefore, many previous studies have focused on pretreatment to improve bioavailability and remove partial ammonia. The most common method is heating and its combination with stripping. The survey by Ardic and Taner [87] found pretreating CHM at 100 °C for two hours improved methane production. Rodriguez-Verde et al. [43] conducted a pretreatment of poultry-pig manure that consisted of temperature simultaneously with ammonia stripping, resulting in a nitrogen removal efficiency of 72% and a 1.2-fold higher methane production. Elasri et al. [88] obtained a biogas yield of 230.58 $mL \cdot g^{-1}$ COD after two-step pretreatments of heating at 105 °C for 24 h following a fine grinding, creating 3–5 times higher than the untreated group. Yin et al. [89] performed a thermophilic pretreatment (70 °C) of CHM, including thermal and ammonia stripping, and found that the methane yield of prehydrolyzed CHM reached 518 mL·g^{-1} VS, which was 54.6% higher than the control reactor. Yin et al. [90] created an innovative two-stage AD by combining thermal stripping pretreatment (70 °C) and an anaerobic membrane bioreactor, bringing a hydrolysis efficiency of 72.4% and methane yield of 352 mL·g^{-1} VS_{in} (growth ≈ 65%).

Additionally, LTPT can accelerate destroying organic molecular bonds in poultry litter by adding chemical reagents. Poultry litter soluble COD increased 2–3 times after conducting a pretreatment of 0.2 g $Ca(OH)_2$·g^{-1} waste at 90 °C, while that at 20 °C showed a wick COD solubilization [42]. Zahan et al. [91] obtained a 45–51% increase of biogas in the co-digesting chicken litter, food waste, and wheat straw after thermal pretreatment at 120 °C with 5% NaOH or 3% H_2SO_4. However, note the inhibitor production of the heat pretreatment of CHM. Both higher temperatures and adding chemicals are more likely to produce inhibitors. Raju et al. [68] found no significant change in the BMP of the CHM after pre-treatment at temperatures up to 200 °C. The use of sodium hydroxide is more likely to produce inhibitors (e.g., volatile fatty acid (VFA), ammonia, furfural) than using lame, and the cations Na^+ and K^+ were potent methanogenic inhibitors when compared with Ca^{2+} [89].

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