

A Review on the Research of Promoting Biogas Fermentation Efficiency by Mixing

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Abstract: Mixed agitation can significantly improve the efficiency of anaerobic fermentation, which is shown by the remarkable improvement of biogas production and pollutant removal rate, so it has become an indispensable auxiliary process in modern biogas engineering. However, the internal mechanism of how agitation improves the fermentation efficiency is not clear, so it is difficult to find the best agitation form under complex conditions. However, in recent years, more research results have been obtained in this field. The effects of various agitation forms have been continuously verified on different feedstock characteristics under different fermentation and operating conditions, and the mechanism has been gradually clarified. The most important achievement is that in the 1990s it was clear that continuous agitation would impact the fermentation system, so batch agitation is the correct way to improve the fermentation efficiency. Since the 21st century, the latest research has focused on setting the operating parameters of batch agitation. Researchers have made greatly progress in all the aspects of medium, mode, power, speed and duration of agitation, and optimized the mixing scheme. Some studies have also revealed that stirring will affect the temperature field, impurity removal, toxicity accumulation and so on, and the optimal design of mixing scheme should take into account the comprehensive effects of all aspects. In this paper, the main worldwide research achievements in this field made in recent years are reviewed, and the internal mechanism that agitation can improve the anaerobic fermentation efficiency of biogas is clarified to a certain extent, and the suitable agitation forms under certain conditions are suggested.

Keywords: Biogas, Fermentation Efficiency, Mixing, Agitation, Flow Pattern

1. Introduction

Under many operating conditions, agitation can improve the efficiency of anaerobic fermentation, and it is presented as the improvement of biogas production and pollutant removal rate macroscopically [1-2]. Nowadays, the low efficiency of static fermentation holds back the development of biogas industry. Generally, biogas projects are installed with agitation devices to increase biogas production rate and pollutant removal rate [3]. However, as the intrinsic mechanism by which agitation can improve the efficiency of biogas fermentation is not clear enough, most studies are restricted to

the speculation based on the surface phenomena [4-5], and some scholars even obtain some contradictory conclusions [6]. Without the support of basic theories, designers have no way to optimize agitation modes and can only add some agitation measures freely, but actually, these are not applicable for digesters and feedstock properties [7]. Poor agitation processes are less effective to improve fermentation efficiency, and more energy-costly [8]. Therefore, the biogas industry calls for more progress in this aspect. With the rapid development of biogas industry, especially the vigorous development of large-scale biogas projects, many countries have replaced traditional household biogas digesters with large-scale biogas projects [9-11], which puts forward higher

requirements for the design of agitation process [12-13]. The large-scale biogas plants such as Deqingyuan Ecological Farm were constructed, with the annual treatment capacity of 80,000 t of chicken manure and 100,000 t of sewage [14]. Without a good agitation scheme, the slurry can easily form silt and the whole system will stop running.

Nevertheless, before the intrinsic mechanism by which agitation can improve the fermentation efficiency has been completely known, studies may not only reveal the action mechanism by which agitation improves the fermentation efficiency in certain particular aspects [15], but also help optimize the design in some particular aspects as reference especially under particular operating conditions [16]. At present, the biogas industry is facing great development opportunities [17-18]. This paper reviews the current research progress of agitation in the field of biogas, hoping to help meet the requirements of the biogas industry for the continuous improvement of the agitation process.

2. Review on the Research of the Fermentation Slurry Agitation

2.1. Knowledge of Agitation

With regard to the influence of agitation on the anaerobic fermentation of biogas, the understanding of the academic community has experienced a tortuous process. In fact, before the 1990s, people did not believe that agitation could increase biogas production rate. On the contrary, people believed that agitation was a kind of impact on the fermentation liquid and would reduce the biogas production [19-20]. There are many explanations for this: for example, Yang Baolin believed that anaerobic digestion is a continuous and stable process, and the input of external power will interrupt this process [21]. M Zaiat believes that agitation will destroy the shape of the sludge granules in the fermentation liquid and result in fragmentation thereof, which is not conducive to fermentation [22]. By 2001, Jarvis P still believed that, at least during the start-up phase, agitation would destroy the fragile surface flocculation structure of biomass, which is not conducive to start-up [23]. Later, people gradually realized that the case of reducing the biogas production rate by agitation was only due to incorrect specific way of agitation. For example, continuous agitation is excessive agitation actually. If proper agitation can be adopted, for example, appropriate intermittent agitation, the biogas production rate can be improved. After the 1990s, it has been widely recognized in the academic community that suitable agitation can significantly improve the efficiency of anaerobic fermentation, and it can significantly improve the biogas production rate and pollutant removal rate macroscopically [1-2].

2.2. Study on the Agitation Modes

Under the premise that scientific agitation is conducive to improving the efficiency of anaerobic fermentation and thus increasing the biogas production rate and pollutant removal

rate, since the 1990s, people have conducted in-depth study on how to scientifically develop the agitation modes. The specific agitation modes are mainly studied from the following three aspects [24].

First, selection of agitation media. The agitation media mainly includes three categories: mechanical agitation (blade agitation, impeller agitation), hydraulic agitation (jet agitation, digestate reflux agitation, slurry circulation agitation) and pneumatic agitation (biogas reflux agitation). With regard to the advantages and disadvantages of the three categories of agitation media, in fact, there are many disputes in the academic community, and so far there is no particularly authoritative conclusion. Only in terms of the effect of improving the gas production rate, the three categories of agitation media are all considered to have the greatest effect of improving the gas production rate, and the ideas are respectively proved by experiments [25-27]. In addition to improvement of the biogas production rate, some scholars discussed the advantages and disadvantages of the three categories of agitation media in terms of systematics, stability and other aspects. For example, Karim pointed out that although hydraulic agitation improves the biogas production rate most, the hydraulic retention time is the shortest, which will cause decrease of the methane content [28]. Borole pointed out that different agitation media gives different success rate of start-up, and only the success rate of hydraulic agitation can reach 100% [29]. Naomichi Nishio pointed out that the dominant methanogen varies with agitation methods, so the applicable agitation method should be determined according to the main species of methanogens in the fermentation liquid [30].

Second, frequency of agitation. As late as the early 1980s, the academic community still believed that agitation should be continuous [31]. It was till 1985 that Ben-Hasson clearly stated that agitation was necessary, but too frequent agitation was not good for biogas production rate, so intermittent agitation should be performed at a suitable frequency [32], which eventually put an end to the current argument over whether manual agitation would increase or decrease gas production rate. Qin Feng provided an important reference for the frequency of agitation through experiments—agitation every 4 hours [33]. But, Qin Feng gave this conclusion only based on experimental phenomena, and did not explain in detail in terms of the mechanism why this frequency is the most appropriate.

Third, input power of agitation. Since agitation itself is energy consuming, if the gas production improved by agitation is not enough to make up the energy consumed by agitation itself, the agitation will be considered to be unworthy. Some algorithms believe that the energy consumed by agitation can account for about 50% of the operational energy consumption of the entire biogas project [34]. But in fact, there are few research achievements in this aspect. One of the important reasons for this is that in the 21st century, a new common view has gradually formed in the modern biogas industry that the main goals of biogas project are environment protection and pollution control, rather than energy capture, so

its own energy consumption is not so important [35-39]. Especially after the industry has determined to adopt low-intensity intermittent agitation, the energy consumed by the agitation is actually very low, so this issue is no longer the primary focus [40].

2.3. Amplitude of Fermentation Efficiency Improved by Agitation

On the basis of determining that suitable agitation can increase fermentation efficiency, the issue that researchers and designers most concerned about is the amplitude that the fermentation efficiency can be improved, and conclusions about this issue from scholars of various countries are very different.

Yakahiro Hiraoka used the residual sludge from the sewage treatment plant for fermentation and adopted mesophilic fermentation, and it was found that biogas yield was $160 \text{ mL}\cdot\text{gCOD}^{-1}$ without agitation and could be increased to $224 \text{ mL}\cdot\text{gCOD}^{-1}$ with agitation, with an increase rate as high as 40% [41]! It shows that the effect of agitation to increase biogas production of anaerobic fermentation is more remarkable under mesophilic fermentation conditions. However Ryuki Enta employed the same raw material for fermentation as Pingwang Zhengsheng, but conducted contrast experiments under different concentration and dilution conditions and found that with a high VS load of 4.7 to $5.9 \text{ kg}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$, the higher the frequency, the higher the gas production rate; but with low VS load below $2.4 \text{ kg}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$, the effect of agitation frequency on gas production is not significant [41].

Youngsukkasem used carboxymethyl cellulose (CMC) and sodium salt as additives and mixed them up to increase the growth rate of methanogens, thereby greatly increasing the gas production rate [42]. Klanarong Sriroth used local cassava waste as raw material, and performed anaerobic fermentation after mixing with sludge to obtain a high biogas production rate [43]. Hopfiner-Sixt pointed out that the increase of biogas production rate is proportional to the degree of material mixing [24]. Stephan Tait used barley straws and concentrated sludge from pigsty to perform mixed fermentation and achieves 60% degradation rate of solid materials [44]. Krzystek found that a low concentration of domestic sewage can be re-introduced into the digester after an anaerobic reaction process, which can increase the biogas production rate 4 folds [45]. Lehtomäki used cow dung and wheat straws to perform mixed fermentation and found that when the ratio of feces and straws was 10:90, the methane content was 16%; when the ratio was adjusted to 25:75, the methane content was still 16%; when the ratio was adjusted to 50:50, the methane content dramatically increased to 31% [46]. Kristian Fjørtoft studied the local biogas fermentation in the cold season and quantitatively described that in cold countries like Norway, the biogas production in the cold season is not always very low, but it can greatly fluctuate between 26.9% and 88.2% of the average biogas production in warm season, thus it is very unstable [47]. Martí-Herrero designed a solar greenhouse for this, and used plastic rings to set grills in a biogas digester to optimize the flow field for reflux and agitation, increased the

biogas production rate by 44%, allowed the biogas digester to operate smoothly at an extremely low temperature of 6.1°C , and improved the operating ability of biogas digesters in low temperature regions in an extremely inexpensive manner. Sang Rak Lee experimentally demonstrated that gas pressure is increased linearly in pneumatic agitation, but the volumetric gas producing rate can only reach $1.25 \text{ L}\cdot\text{L}^{-1}\cdot\text{d}^{-1}$ at most, and no longer increases thereafter [49].

Different scholars obtained different amplitudes of improvement. It can be seen that the effects of agitation under different conditions are quite different. This also suggests that different agitation designs must be used under different conditions to achieve improvement effect of greater amplitude.

3. Research Progress on Mechanism of Agitation to Improve Fermentation Efficiency

3.1. Agitation Can Promote Thorough Broth Mixing

The most intuitive reason why agitation can improve the fermentation efficiency is that agitation can mix the materials evenly and promote sufficient reaction. Many scholars have pointed out that the mixed fermentation of various materials, such as feces, straws, waste papers, and kitchen waste, is often more efficient than the separate fermentation of each material [50-59], and mixed fermentation of multi-strains can also improve the methane conversion rate [60-61], but it is even more necessary to enhance the mixing and agitation, so that a variety of materials are mixed evenly.

Li Shulan found that scum can be produced in upper, middle and lower parts of the anaerobic tank using straws as raw material, but the potential of gas production is only $0.08 \text{ mL}\cdot\text{g}^{-1}$, $0.16 \text{ mL}\cdot\text{g}^{-1}$ and $0.23 \text{ mL}\cdot\text{g}^{-1}$ respectively, which is far lower than that of direct fermentation [62], so with agitation, fermentation feedstock can be well mixed to increase the total fermentation efficiency. Li Jiang believes that in addition to main nutrients for growth of anaerobic methanogens such as nitrogen (N), phosphorus (P) and sulfur (S), the slurry also contains trace metal elements such as potassium (K), iron (Fe), cobalt (Co) and nickel (Ni) which are also essential nutrients for their growth, but these elements have very low content in the slurry [63]. In large digesters, they must be stirred to diffuse into large space. Wang Yuheng observed the shape of the flocculates of the solid sludge in the hydraulic agitation using a microphotographing system, and believed that the cutting force of agitation destroys the loose structure on the surface of the solids, exposing dense compact parts and making the granules solid particles smaller averagely, so thorough contact with the sewage is realized, thereby improving the fermentation efficiency. Wang Yuheng further believes that agitation should also be combined with the chemicals to enhance the fermentation efficiency [64]. Li Yang used a long-cylinder anaerobic fermentation tank with a height of 2800 mm and a diameter of 50 mm to carry out

anaerobic fermentation experiments on pig manure, and found that after several hours of gravity sedimentation, the feedstock is separated into two forms: concentrated sludge and diluted liquid. The concentrated effluent, diluted effluent and unseparated slurry were used as feedstock respectively to conduct experiments on independent gas production from anaerobic fermentation. Regarding the gas production efficiency, it can be ranked as: concentrated sludge > fresh wastewater > diluted slurry, and the difference was significant [65], so it is necessary to fill more areas with concentrated sludge, reducing diluted slurry by agitation. Whereas Luo Tao found that in long-cylinder anaerobic fermentation tanks, if the feedstock is fed from the top, and it is slowly settled under the function of gravity, the fermentation efficiency during the sedimentation process is excellent [66]. Based on this, Huang Ruyi believes that the intrinsic mechanism by which agitation can improve the efficiency of fermentation is that agitation can make the slurry resistant to gravity and maintain a state of suspending in the liquid column and slowly sediment over a long period of time [67].

3.2. Influence of Agitation on Biofilm

Some scholars have tried to explain the mechanism by which agitation improves the efficiency of anaerobic fermentation from the view of biofilm. Liu Jingtao believes that anaerobic fermentation treatment of wastewater is beneficial to stable mass transfer by forming biofilm in the wastewater by the solid carrier, and made comparison on the biofilm formation efficiency of three biofilm carriers [68]. Zhao Qingliang and Huang Yuchang often added waste tire particles as carrier for biofilm formation on the basis of traditional active sludge process, which can increase the biofilm amount by 50% to 100% on the basis of traditional process [69]. The main role of the biofilm is to filter out some useless oil and other substances to ensure high-efficiency anaerobic fermentation [70]. But, Yang Ping studied the process of wastewater treatment by anaerobic biological fluidized bed with polymer porous carriers and found that thick biofilm can also hinder mass transfer, but agitation can just make thin the biofilm [71]. At the same time, Yang Ping also corrected Tavares's view that the faster the flow rate of agitation under multiphase flow conditions, the thinner and the more dense the biofilm [61], because the hydraulic shear force only needs to reach a critical value, the biofilm begins to fall off.

3.3. Influence of Agitation on Temperature Field

Some scholars have also studied the influence of agitation on the temperature field. The academic community has always accepted that warming can promote the anaerobic fermentation of biogas [73], but if agitation is conducted at the same time, the temperature field will be affected [74]. Shi Huixian pointed out that if an anaerobic fermentation system uses both agitation and warming processes, agitation will cause spatial heterogeneity in the fluid temperature field inside the reactor. From view of fermentation temperature, it is

not conducive to anaerobic fermentation reaction [75]. Bi Junwei found that the influences of different agitation rates on the temperature field are quite different and cannot be ignored [76]. Li Daoyi found that continuous agitation at high temperature (55 °C) can not increase the gas production rate, instead, intermittent agitation can decrease the gas production rate by about 10% [77]. It can be seen that under the warming fermentation condition, agitation measures need to be carefully added.

3.4. Agitation Can Inhibit Toxicity Accumulation

During the anaerobic fermentation of biogas, some toxic substances are produced and accumulated continuously. Agitation can inhibit toxicity accumulation and protect generation reproduction of methanogens. This is probably one of the important mechanisms by which agitation can improve fermentation efficiency.

Hu Ping found in experiments that during the mixed anaerobic fermentation of cyanobacteria and sludge, cyanobacteria would release toxins and inhibit fermentation [78]. Xu Lijuan found that fresh cyanobacteria can be decomposed and then perform anaerobic fermentation to produce biogas, without toxicity appearing at the initial stage. However, after 15 days of gas production, toxicity will still appear with the recovery of amylase and dehydrogenase [79]. Based on the Yakahiro Hiraoka [41], Zhang Liguang experimentally demonstrated that the dominant methanogens in the up-flow anaerobic sludge bed (UASB) system are *Methanosaeta concilii* and *Methanospirillum hungatei*, while the dominant methanogens in the continuous-flow stirred tank reactor (CSTR) system were *Methanosarcina mazei* and *Methanobacterium formicicum*. It was clearly pointed out that the differences in composition and metabolic characteristics of the microbial communities in the sludge are the internal causes of differences in the efficiency of anaerobic fermentation systems, whereas different agitation modes have different effects on toxin release of floras and provide important reference for how to select the agitation mode for different systems [80].

The anaerobic fermentation of biogas continuously produces volatile fatty acids, which are the most important precursors of methane, and also have strong inhibitory effect on the generation reproduction of methanogens [81]. This is often reflected in the acidification of the broth, i.e. pH decreases, and agitation has great influence on this [82]. Duan Xiaorui found that the more intense the agitation, the more favorable to the production of organic acids [83]. But on the contrary, this is also prone to cause acidification of the broth, which is not conducive to generation reproduction of methanogens, so there is an urgent need to find the most appropriate agitation strength. Du Lianzhu also found the same laws, compared the acid production effects of pig manure and straw materials mixed at different proportions, and believed that when the mass ratio of pig manure to straw was 2:1, the proportion of acetic acid in produced fatty acids was the highest, indicating that the same agitation mode has inconsistent effects on raw material of different mixing status

[84]. Chen Jiajia's viewpoint is contrary to them, he believes that in the anaerobic fermentation process with straw as the only raw material, the agitation rate cannot be too low, otherwise it is prone to cause acid accumulation and inhibit fermentation efficiency [85-86]. Differences therebetween may be related to the specific kinds of raw materials.

The more important toxic substance is ammonia nitrogen. Carl Hansen pointed out that during the anaerobic fermentation of biogas, ammonia nitrogen is continuously produced and accumulated. Ammonia nitrogen has a double-edged-sword effect on anaerobic fermentation of biogas. It is not only a food for methanogens, but excessively high ammonia nitrogen concentration has toxic effect on the reproduction of methanogens. Whereas agitation can inhibit the accumulation of ammonia nitrogen and reduce the concentration of ammonia nitrogen, so it is conducive to the growth and generation reproduction of methanogens, and improves the fermentation efficiency. Carl Hansen also established a four-stage ammonia nitrogen accumulation model for anaerobic fermentation [87]:

Stage 1: $0 < [NH_3] < 1.10$, $\mu_r = 1.0$

Stage 2: $1.10 < [NH_3] < 1.16$, $\mu_r = \frac{1}{-7.6 + \frac{[NH_3]}{0.128}}$

Stage 3: $1.16 < [NH_3] < 1.34$, $\mu_r = 0.67$

Stage 4: $1.34 < [NH_3]$, $\mu_r = \frac{1}{-12 + \frac{[NH_3]}{0.995}}$

The Hansen model is of great significance for explaining the fundamental mechanism by which agitation can improve the gas production rate and guiding targeted agitation. On this basis, He Shijun has defined the boundary of the effect of ammonia nitrogen on the activity and toxicity of methanogens through experiments: $800 \text{ mg} \cdot \text{L}^{-1}$ [88]. However, Zhao Qingliang and Li Xiangzhong believe that the concentration of ammonia nitrogen will rapidly double from $50 \text{ mg} \cdot \text{L}^{-1}$ to this critical value, so the concentration of ammonia nitrogen must be reduced to below $50 \text{ mg} \cdot \text{L}^{-1}$ before fermentation starts up safely [89]. However, Yu Fangfang and Wu Jiandong believe that under the operating condition with different carbon sources, this boundary is also different and cannot be generalized [90]. Li Yafeng also believes that under the operating condition with higher COD concentration, sufficient carbon sources can inhibit production of ammonia nitrogen [91]. Zhang Bo from Shanghai Jiaotong University proposed that the process of ammonia nitrogen accumulation is reversible. It can reduce the concentration of ammonia nitrogen and restore the activity of methanogens through influent water (i.e., hydraulic agitation) [92]. Jia Chuanxing has established a more complete two-phase anaerobic digestion and ammonia nitrogen accumulation model based on the four-stage ammonia-nitrogen accumulation model of Carl Hansen, and Jia Chuanxing pointed out that the reflux ratio of digestive fluid (i.e., the flow rate of hydraulic agitation) is the key parameter to control ammonia nitrogen accumulation [93].

Gao Yanning believes that circulating fluidization is a better method of removing ammonia nitrogen than

mechanical agitation [94]. He designed a complex circulating fluidized bed anaerobic ammonia oxidation reactor and established a convection-diffusion-response coupling transport equation, obtained the distribution rule of sludge concentration along the height of the reactor under different operating parameters [95], and proposed that the most suitable reflux ratio of the complex circulating fluidized bed anaerobic ammonia oxidation reactor is 200% to 300% [96]. But Guo Yong believes that ammonia nitrogen can be removed by using anaerobic ammonia oxidation denitrification method without stirring [97]. Sui Jichao and Jiang Jianguo also pointed out that the anaerobic fermentation process itself consumes a lot of ammonia nitrogen, and in most cases only during the start-up phase the concentration of ammonia nitrogen is very high and then naturally decreases [98]. Moreover, the critical value of ammonia nitrogen toxicity under most operating conditions is very high and is not easy to achieve under normal conditions, so there is no need to add agitation [99].

4. Conclusion

Mixing and agitation can handle more complex raw material conditions and can also achieve better fermentation efficiency, and have become an indispensable process type for modern biogas projects. Although the fundamental reason why agitation can increase the efficiency of biogas fermentation has not yet been completely clarified, many results have illustrated the improvement effect of agitation in certain particular aspects and can be used as theoretical guidance for design of agitation processes under these particular conditions. Many misunderstandings about agitation have been discerned, making the design of modern processes more reasonable. Especially in recent years, there have been many research results in terms of inhibiting the toxicity accumulation by agitation. It has been a good guide to optimize the design of agitation parameters by monitoring the content of toxic substances in the sludge, which has greatly promoted the development of biogas industry.

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