

Article

Using Chemically Enhanced Primary Treatment (CEPT) as a Pretreatment Option for Anaerobic Digestate from Cattle Manure Digestion System

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Abstract: Anaerobic digestate from cattle manure fermentation may pose a threat to the environment. How to stabilize the digestate's characteristics so that they do not disturb the bio-system is a critical issue for digestate management. Chemically enhanced primary treatment (CEPT) was investigated as a pretreatment option for digestate treatment. The performance of CEPT for digestate management was carried out under rapid mixing (200 r/min) and slow stirring (40 r/min), respectively. The optimal dosage of ferric chloride (FeCl_3) was 40 mg/L. The combination of FeCl_3 and anionic polyacrylamide (APAM) had no obvious influence on TP removal, while COD removal efficiency was improved by 15.4%. The digestate pH and temperature remained stable for CEPT application and required no adjustment. The results indicate that CEPT was effective and feasible in enhancing the removal of COD and TP for digestate pretreatment by using FeCl_3 and APAM.

Keywords: anaerobic digestion (AD); digestate; chemically enhanced primary treatment (CEPT); pretreatment; ferric chloride

1. Introduction

In recent years, the handling and recycling of animal manure and organic wastes has received considerable attention [1]. Using animal manure and organic wastes as feedstock means that renewable energy systems create environmental, energy, and economic benefits simultaneously such as greenhouse gas (GHG) reduction, clean energy supply, and cost savings [2]. Wastes can be converted into different and useful forms of energy through various technologies and processes. One promising process for biological conversion of organic wastes is anaerobic digestion (AD), which turned wastes into renewable energy and biofuel in the form of methane. This method is arousing more interest because it has the ability to mitigate the impact of wastes on the environment and facilitate sustainable development [3,4]. AD is an effective biochemical process for organic waste treatment and for biogas production because AD offers numerous significant advantages, such as low sludge production, low energy requirement, and green energy recovery. It has been evaluated as one of the most energy-efficient and environmentally beneficial technologies for bioenergy production [5–7].

AD or co-digestion of organic wastes has been extensively researched and demonstrated. Although the AD process for organic waste treatment is very complicated and requires further

analysis before it is employed in commercial applications, it has become the default process for biological conversion of organic wastes into renewable energy in the form of methane. The application of AD for the treatment of organic wastes has made spectacular advances in recent years [3,8,9]. On the other hand, an organic matter-rich solid waste is generated after the AD process, which is defined as digestate [10]. The properties of digestate are highly dependent on the properties of the feedstock used [11]. Applying digestate as fertilizer was once considered to be the most suitable way to enrich nutrients, and its quality must satisfy the needs of nations' regulatory frameworks [12]. Due to the increasingly strict regulations on its contents of heavy metals, pathogens, and organic pollutants, concerns regarding anaerobic digestate's appropriate disposal, treatment, and reuse have increased [12,13]. In addition, the feasibility of its direct fertilization of soil will be constrained by transport requirements.

Having many large-scale biogas plants operating may lead to an oversupply of digestate in certain regions. If the use of digestate is limited by agricultural area, the surplus has to be transported elsewhere, meaning that transportation costs and logistical problems have to be considered [14]. If the digestate has to be discharged into the environment, the post-treatments essential for AD alone may lead to effluent failing to meet discharge standard requirements as used in most industrialized countries. Based on an assessment of the current status of digestate treatment technologies, the applied treatment concepts can be classified into two approaches: partial treatment or complete purification [15]. Generally, partial treatment aims mainly to reduce quantity or serve as a pretreatment unit for subsequent processes. The complete purification process mainly concentrates on valuable nutrients and advanced purification for safe discharge.

Membrane technologies have proven effective for the advanced treatment of digestate [15–17]. Membrane technologies can assist in the concentration of purification in two ways. Concentration usually plays an important role in making digestate transportation economically feasible and beneficial as a fertilizer [18,19]. For areas where digestate is not available for agricultural purposes during cold seasons, purification can help to protect the environment by removing pollutants effectively [15,20–22]. It has been reported that anoxic/oxic-membrane bioreactor (A/O-MBR) was technically feasible for the treatment of digestate, and can provide an environmentally acceptable way for the application of wet-AD in animal manure treatment [16]. However, it was found that wide variations of digestate quality could affect A/O-MBR efficiencies adversely during the year. The contents of digestate are often varied: firstly, according to the composition of feedstock digested; and secondly, over time for the same AD plant [15]. How to provide a stable and good quality digestate for the A/O-MBR system has become a critical issue in full-scale biogas plants. If high-quality AD feedstock cannot be guaranteed all the time, effective pretreatment for the subsequent system may be a better solution to stabilize the quality of influent for biological treatment processes.

Chemically enhanced primary treatment (CEPT) is a process that has been known for more than 100 years. However, its application to wastewater was consolidated only in the past two decades based on technological development of coagulation [23]. CEPT can remove suspended solids (SS), colloids, and total phosphorus (TP) effectively. According to the literatures, CEPT can remove SS (90%), BOD (50–70%), COD (50–60%), and TP (80–90%) [23–27]. Furthermore, CEPT possesses many other advantages, such as reducing the footprint of pretreatment unit, reducing the cost, low energy requirement, easy to operation and maintenance, etc. [26]. However, CEPT is not very effective in removing ammonium and dissolved organics, which limits the application of CEPT [25,27]. Little work has been reported on the application of coagulants with cationic polymers for CEPT of digestate. The objective of this study was to evaluate the effectiveness of CEPT as a pretreatment option for anaerobic digestate from a cattle manure digestion system. Based on the efficiency in removing COD and TP, the CEPT process was optimized to achieve the most suitable parameters for ensuring the effluent was of a quality suitable for A/O-MBR.

2. Materials and Methods

2.1. Experimental

The literature has documented that residual alum is harmful to human health and iron coagulants are more suitable for application in CEPT [28,29]. According to the influent's qualities and former results on wastewater treatment, ferric chloride (FeCl_3) was selected for its popularity and easy-to use features, and anionic polyacrylamide (APAM) was used as a coagulant aid.

2.2. Wastewater Characteristics

The digestate was extracted from the supernatant after 24.0 h sedimentation of effluent being discharged from AD treatment of cow manure. The main parameters of the digestate used in this study are shown in Table 1.

Table 1. Characteristics of the digestate used.

Parameter	Average Value	Minimum Value	Maximum Value
pH	7.31	7.03	7.78
COD (mg/L)	4352	2687	5088
TP (mg/L)	132	107	158

2.3. Sampling and Analytical Methods

Measurements of the conventional parameters such as pH, COD, and TP were conducted according to the standard methods [30]. The chemical oxygen demand (COD) concentration was measured through the rapid digested spectrophotometry method using potassium dichromate with a microwave digestion system (XJ-III, Taihong medical instrument Co., Ltd., Shaoguan, China). The concentrations of TP were determined using a UV-Vis spectrometer (UV-8000S, Shanghai Metash instrument Co., Ltd., Shanghai, China) at wavelengths of 700 nm. The pH was measured using a pH meter (PB-21, Sartorius, Goettingen, Germany), which was calibrated weekly using commercial buffer solutions.

2.4. Experimental Procedure

In our experiment, 1 L beaker was used as container. MY3000-6M (Wuhan Meiyuyiqi Co., Ltd., Wuhan, China) six-link coagulation test agitator apparatus was also employed in CEPT tests. In order to ensure the analysis of the quantity of water samples, 6 L biogas was used in each test group, and the supernatant of $400 \text{ mL} \times 6$ was used for measurement after the static precipitation. Jar tests were conducted with rapid mixing for 1 min, slow stirring for 10 min, and sedimentation for 15 min. FeCl_3 was dosed when rapid mixing began and 5 min later PAM was dosed (during combined use). From past experience, several different stirring speeds were compared to achieve different coagulation strengths. Different conditions used in the experiments are shown in Table 2. After sedimentation, the supernatants were sucked using a pipette to measure COD and TP values. The chemical oxygen demand (COD) total phosphorus (TP) removal efficiency was calculated by Equations (1) and (2)

$$\text{COD}_{\text{removed}}(\%) = \frac{\text{COD}_{\text{influent}} - \text{COD}_{\text{effluent}}}{\text{COD}_{\text{influent}}} \times 100 \quad (1)$$

$$\text{TP}_{\text{removed}}(\%) = \frac{\text{TP}_{\text{influent}} - \text{TP}_{\text{effluent}}}{\text{TP}_{\text{influent}}} \times 100 \quad (2)$$

where $\text{COD}_{\text{influent}}$ and $\text{TP}_{\text{influent}}$ were the COD and TP in the extracted digestate, respectively. $\text{COD}_{\text{effluent}}$ and $\text{TP}_{\text{effluent}}$ is the COD and TP concentration in the effluent after CEPT, respectively.

Table 2. Experimental conditions.

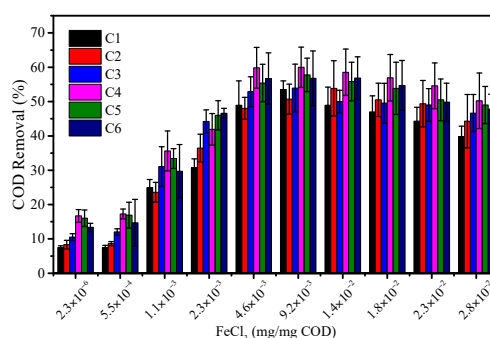
Condition	C1	C2	C3	C4	C5	C6
Rapid mixing (r/min)	300	200	300	200	300	200
Slow stirring (r/min)	30	30	40	40	50	50

3. Results

3.1. Effect of Conditions and Dosages on CEPT's Efficiencies

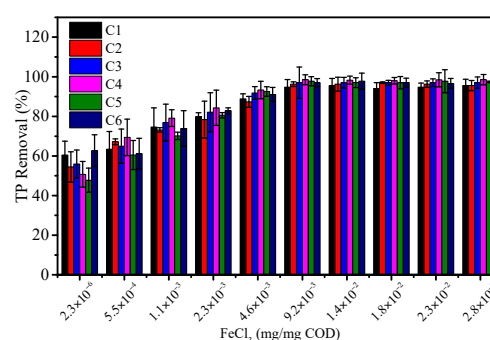
3.1.1. COD Removal

The dosing of FeCl_3 could help to remove COD, and the removal efficiencies increased with incremental dosages under different conditions. It is shown in Figure 1 that removing COD by CEPT could be much more efficient at the dosages of 20, 40, and 60 mg/L, respectively. It could be seen that an optimum range existed for coagulant dosing in COD removal. The best COD removal efficiency was achieved using 40 mg/L coagulant, and no obvious enhanced removal efficiencies resulted by increasing the dosage. From Figure 1, it is also evident that the best coagulation scenario was C4 (rapid mixing at 200 r/min, slow stirring at 40 r/min) under different dosages. Furthermore, the most efficient removal of COD was 60% at 40 mg/L, C4.

**Figure 1.** Effect of coagulation on COD removal efficiencies.

3.1.2. TP Removal

According to the literature, CEPT is efficient in removing TP. Figure 2 shows that FeCl_3 can remove TP efficiently under different coagulation conditions. Removal efficiency increased with the dosing and achieved stability at a dosage of 40 mg/L. Also, the C4 improved TP removal efficiency than under other conditions. Given the chemical cost and energy consumption, the best TP removal efficiency was 98.51% at 40 mg/L under C4.

**Figure 2.** Effect of coagulation on TP removal efficiencies.

3.2. Efficiencies of Combined Use of FeCl_3 and APAM in CEPT

The combination of coagulants with anionic polymers as a coagulant aid for CEPT of wastewater also exhibited better removal efficiencies for pollutants [31,32]. The combined uses of anionic polyacrylamide (APAM) with coagulants in wastewater treatment have been reported as resulting in a number of benefits, for example: improving efficiency in removing pollutants while saving coagulants; and producing less sludge volumes with good dewaterability [31–33]. In Figure 3, it was shown that the addition of APAM can increase COD removal efficiencies for CEPT, while no obvious effects were noted for TP removal. The reason should be that FeCl_3 itself can remove TP efficiently and the improving space is limited. With reference to COD removal, 0.1 mg/L APAM could increase COD removal efficiency by 10.2% and 0.2 mg/L APAM could increase it by 15.4%. Table 3 shows that CEPT was effective as a pretreatment in COD and TP removal. The combined use of FeCl_3 and APAM had no obvious effect on TP removal, while COD removal efficiency significantly increased by 15% with 0.2 mg/L APAM addition. However, the COD and TP removal efficiency are still lower than some previous reports due to the special properties of digestate. The complicated chemical composition and high organic content of the digestate could severely affect the performance of CEPT.

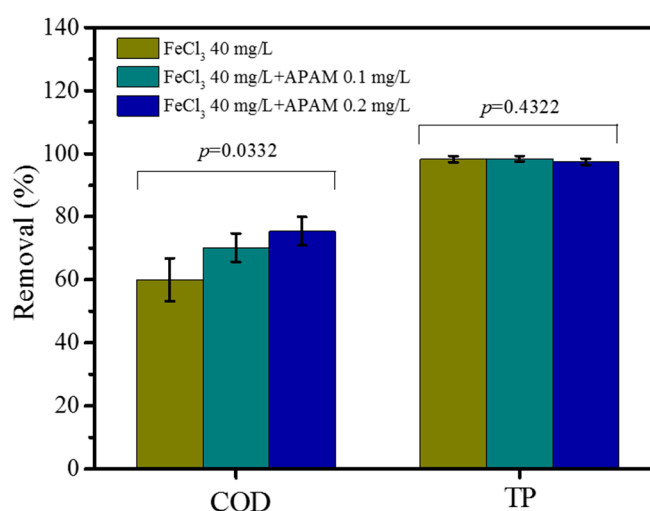


Figure 3. Effect of FeCl_3 + APAM dosing on anaerobic digestate treatment (FeCl_3 = 40 mg/L).

Table 3. The comparison of CEPT's efficiencies.

CEPT	COD (%)	TP (%)	Reference
$\text{Fe}_2(\text{SO}_4)_3$	78	99	[31]
$\text{Fe}_2(\text{SO}_4)_3$ and APAM	94	99	
$\text{Al}_2(\text{SO}_4)_3$	87	99	
$\text{Al}_2(\text{SO}_4)_3$ and APAM	88	99	
HP-PACl	69	78	[26]
FeCl_3	58	90	[34]
FeCl_3	60	98	This study
FeCl_3 and APAM	75	97	

3.3. Factors Influencing CEPT's Efficiencies

The efficiency of CEPT may vary according to changes in temperature, pH, and coagulation conditions [23]. Coagulation conditions have been investigated and compared. The effects of temperature and pH on CEPT's efficiencies are described in more detail below.

Inorganic coagulant's hydrolysis is an endothermic reaction, and therefore low temperatures could affect hydrolysis reaction adversely and impair the CEPT's effectiveness. It was shown that temperature had obvious effects on COD and TP removal (see Figure 4.). The removal efficiency increased by 27.5% (COD) and 36.9% (TP) respectively, when the temperature rose from 5 °C to 25 °C. For the treated digestate from cattle manure fermentation, the methanogenic tank's temperature was retained at 35 ± 1 °C, the digestate's temperature varied from 18 °C to 23 °C. The digestate's temperature range had no apparent impact on CEPT's effectiveness based on the results under different temperatures. Consequently, no measures were needed to optimize temperature for improving CEPT.

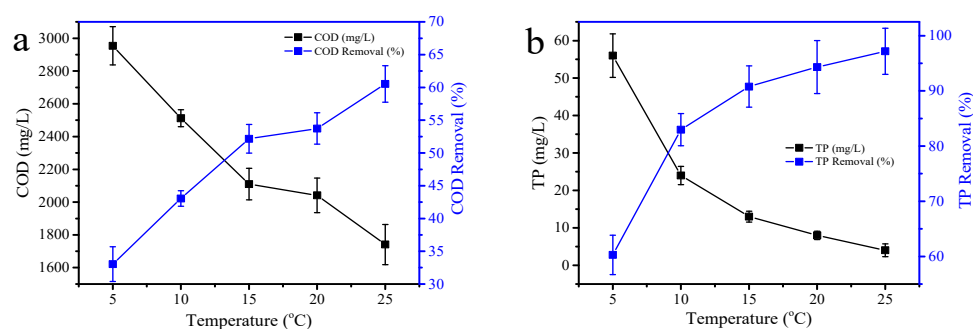


Figure 4. Effect of temperatures on CEPT efficiencies: (a) COD removal and (b) TP removal (FeCl_3 40 mg/L, APAM 0.2 mg/L).

The coagulation process is sensitive to a solution's pH for the hydrolysis and bridge processes are directly related to solution pH. From Figure 5, it is evident that CEPT remained stable in its operation at a pH of 6–9, and the best COD removal efficiency (increasing by 3.7% compared with the lowest) was achieved at pH 7.0. Meanwhile the best TP removal efficiency (increasing by 3.5% compared with the lowest) was achieved at pH 8.0. The digestate's pH varied from 7.03 to 7.78 during the whole experimental phase, and $\text{Fe}(\text{OH})_3$ was reported to be an atypical amphoteric compound that is suitable when the pH ranges from 5 to 10 [25]. Thus, there was no need to adjust pH for optimizing CEPT.

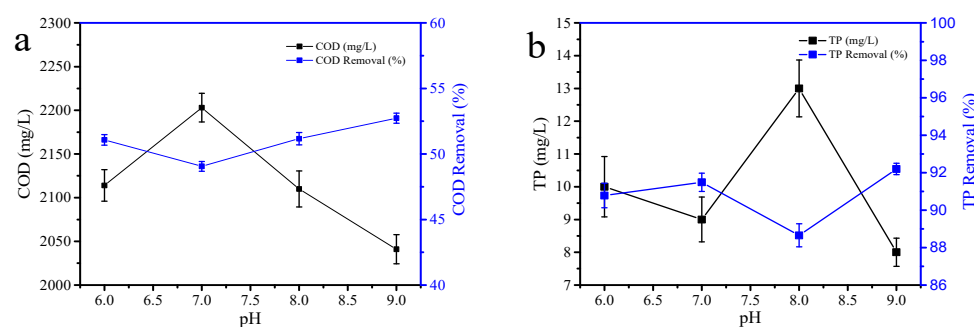


Figure 5. Effect of pH on CEPT efficiencies: (a) COD removal and (b) TP removal (FeCl_3 40 mg/L, APAM 0.2 mg/L).

4. Conclusions

The treatment of anaerobic digestate derived from cattle manure fermentation by means of CEPT using FeCl_3 and APAM was investigated in this study. The experimental results demonstrate that CEPT was effective as a pretreatment in COD and TP removal, which can alleviate the subsequent A/O-MBR's load and improve the effluent's quality. The best coagulation scenario was rapid mixing (200 r/min) and slow stirring (40 r/min) at the best dosage of 40 mg/L FeCl_3 . The combined use of FeCl_3 and APAM had no obvious effect on TP removal, while COD removal efficiency significantly

increased by 15.4% with 0.2 mg/L APAM. Finally, the digestate's pH and temperature remained at a suitable status for CEPT's application and no adjustments were required.

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Conflicts of Interest: The authors declare no conflict of interest.

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