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Peracetic acid enhanced anaerobic co-digestion of excess sludge and food waste: Performance and mechanism

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ABSTRACT

Anaerobic co-digestion of excess sludge (ES) and food waste (FW) has been proven to be a clean and efficient strategy for the resource recovery of organic waste. However, the methane production from the anaerobic co-digestion of ES and FW is not optimal. This study reports a new strategy of using peracetic acid (PAA) pretreatment to enhance the anaerobic co-digestion of ES and FW and reveals the underlying mechanisms. The results confirm that PAA can effectively promote the production of methane from the anaerobic co-digestion of ES and FW, with the maximum methane yield reaching 416 mL/g volatile suspended solids at a PAA content of 9% (w/w). Mechanistic analysis indicates that PAA efficiently facilitates the solubilization of organic matter, promoting the release of soluble proteins and polysaccharides, and accelerating the metabolic conversion of volatile fatty acids (VFAs) to prevent excessive acidification. The maximum output of VFA in the PAA group was 1,511–1,974 mg/L, which was lower than that in the control group. Batch experimental analysis revealed that PAA promoted the hydrolysis and acidification processes, but inhibited the methanogenic process. The PAA pretreatment technique provides a theoretical basis for the efficient treatment of organic matter in urban areas.

Key words: co-digestion, excess sludge, food waste, methane, peracetic acid

HIGHLIGHTS

- Peracetic acid (PAA) enhanced anaerobic co-digestion of sludge and food waste.
- PAA promotes the dissolution of co-digested substrates, thereby ensuring sufficient availability of available substrates.
- The impact of PAA on each step of anaerobic digestion was analyzed.

1. INTRODUCTION

Excess sludge (ES), as a byproduct of sewage biological treatment processes, represents a complex medium characterized by multiple components, media, and interactions (Wei *et al.* 2018). It was reported that the annual output of ES was as high as 30 million tons (Qiang *et al.* 2015). In addition to organic matter, sludge contains a plethora of pollutants such as heavy metals and antibiotics. Improper handling and disposal of sludge can lead to secondary environmental pollution (Cheong *et al.* 2022; Wu *et al.* 2022). Food waste (FW) is also a typical organic waste in urban areas, with an annual production of 600 million tons (Zhang *et al.* 2018). Currently, the main strategies for dealing with ES and FW include incineration, landfilling, composting, and anaerobic digestion (Liew *et al.* 2023; Zhao *et al.* 2023). Anaerobic digestion technology, as a sustainable biological treatment method, offers a viable approach for the decomposition of organic waste and energy recovery, presenting one of the effective methods for ecologically treating sludge (Liew *et al.* 2022). However, the presence of tough cell walls and membranes encapsulating sludge restricts the release and utilization of intracellular substances, thereby resulting in low anaerobic digestion efficiency. Only 30–35% of organic matter within sludge can be converted into volatile fatty acids (VFAs) through anaerobic digestion, leaving the remaining 60–65% unused and requiring further treatment and disposal (Feng *et al.* 2014). Anaerobic digestion of sludge alone suffers from the drawback of carbon-to-nitrogen (C/N) imbalance. Recently, co-digestion of ES with FW has garnered widespread attention, as it offers advantages such as balanced C/N ratios, synergistic disposal of organic waste, and enrichment of microbial communities (Mehariya *et al.* 2018; Zhao *et al.* 2021; Kan *et al.* 2021a, 2022a; Zhang *et al.* 2023a, 2023b).

In order to further enhance the anaerobic co-digestion of ES and FW, various pretreatment technologies, including physical, chemical, and biological methods, have been applied to improve the hydrolysis rate (Yuan *et al.* 2019; Kan *et al.* 2021b; Wang *et al.* 2023; Fu *et al.* 2024; Guan *et al.* 2024). Among these, the addition of chemical reagents offers the advantages of easy operation, high treatment efficiency, and good economic performance. Recently, peracetic acid (PAA) has been widely used for water disinfection, removal of difficult-to-degrade pollutants, and inactivation of pathogens due to its excellent oxidative capacity and bactericidal properties (Kiejza *et al.* 2021; Kan *et al.* 2022b). PAA has a relatively low O–OH bond dissociation energy, which means that when using PAA for water disinfection, the amount of PAA required can be significantly reduced. Ren *et al.* (2024) reported that PAA can enhance the release and transformation of organic matter, and alter the microbial community structure to improve the ES anaerobic digestion efficiency. Additionally, Appels *et al.* (2011) applied PAA to enhance organic hydrolysis to increase biogas production, with the maximum increase in biogas production reaching 21%, however, high doses of PAA led to a decrease in biogas production. PAA has also been used in the dark fermentation of sludge to produce hydrogen, where it can change the fermentation type from propionic to acetic and butyric acids (Li *et al.* 2022). The hydroxyl and acetate radicals produced during the PAA degradation process can significantly improve the conductivity of sludge and suppress the generation of odorous gases during the sludge dewatering process. Based on these findings, PAA can serve as an intensifier to enhance the hydrolysis of organic matter, thereby increasing the resource utilization rate of organic matter. However, studies on the use of PAA to enhance the co-digestion of ES and FW are scarce, and the underlying mechanisms are not well understood. Moreover, it is well-known that the anaerobic digestion process of organic matter involves sequential biochemical stages such as hydrolysis, acidification, and methanogenesis, and how PAA affects these processes is still unclear. Therefore, it is necessary to investigate the impact of PAA pretreatment on the anaerobic digestion of ES and FW to improve the efficient utilization rate of organic solid waste in urban areas.

Based on the aforementioned discussion, this work investigated the impact of PAA on the anaerobic co-digestion of sludge and FW, and revealed the related mechanisms. Firstly, the influence of PAA concentration on the co-digestion of ES and FW was systematically explored. Secondly, the characteristics of organic matter transformation during the anaerobic co-digestion process of ES and FW in the presence of PAA were analyzed. Thirdly, the impact of PAA on each step of the anaerobic digestion process was examined. Finally, the significance of energy recovery based on PAA enhanced anaerobic co-digestion of ES and FW is discussed. The results of the work enrich the application scope of PAA and provide a theoretical basis for the resource recovery of urban organic solid waste.

2. MATERIALS AND METHODS

2.1. Source and characteristics of experimental materials

The ES was sourced from the secondary sedimentation tank of a wastewater treatment plant. The retrieved ES was filtered to remove impurities and was kept on standby for use, with the main characteristics of the experimental ES presented in Table 1.

The FW utilized in the experiments was collected from a university student cafeteria. After collection, the FW was manually screened to remove non-degradable impurities such as chopsticks, plastic bags, and animal bones. It was then pulverized into a paste-like substance using a high-speed blender (Joyoung brand), with the main characteristics of the FW also displayed in Table 1.

Table 1 | Main physicochemical properties of digestive substrates and inoculum used in the experiment

Index	Unit	ES	FW	Inoculum
pH	/	7.1 ± 0.2	6.9 ± 0.1	7.1 ± 0.1
TSS	g/L	4.2 ± 0.3	7.4 ± 0.1	3.2 ± 0.3
VSS	g/L	3.5 ± 0.2	5.4 ± 0.3	2.9 ± 0.2
SCOD	mg/L	210 ± 16	334 ± 15	124 ± 5.3
SPN	mg/L	80 ± 5.3	104 ± 0.2	41 ± 2.3
SPS	mg/L	46 ± 3.2	210 ± 5.9	39 ± 3.9

Note: The data in the table represent the average and standard deviation of the three measurements.

The inoculum was derived from a continuous-flow anaerobic digester in a laboratory, which is primarily used for sludge anaerobic digestion experiments. Upon retrieval, the inoculum was allowed to settle for 24 h to remove the supernatant before being prepared for use, with the main properties of the inoculum showcased in Table 1.

PAA was purchased from a chemical reagent company in Shanghai and stored in a brown bottle in the laboratory upon arrival. The main characteristics of PAA are as follows: Boiling point: 105 °C, relative density (water = 1): 1.15 (at 20 °C), saturated vapor pressure: 2.67 kPa (at 25 °C), flash point: 41 °C. A schematic diagram of the anaerobic SBR device used in this work is presented in the Supplementary material.

2.2. PAA enhanced anaerobic co-digestion of ES and FW

This experiment was conducted in five sets of identical reactors, with each set comprising three identical reactors. Initially, each reactor was fed with 3.0 L of inoculum and 3.0 L of digestion substrate (the ES and FW were mixed at a mass ratio of 1:1 calculated based on total suspended solids (TSS) quality). Secondly, different amounts of PAA were introduced into each group, controlling the mass concentration to be 0, 3, 6, 9, and 12%, respectively (calculated based on the TSS of the digestion substrate). Thirdly, after the addition of materials to each reactor was complete, 2.0 M hydrochloric acid or 3.0 M sodium hydroxide was manually added to adjust the initial pH to 7.0, with no further intervention during the reaction process. Finally, high purity (over 99.99%) nitrogen gas was introduced into each group for 1.0 min to purge the oxygen from the reactors, ensuring a strictly anaerobic environment. The reactors were then quickly sealed with rubber stoppers and transferred to a constant temperature shaker for mesophilic anaerobic digestion (35 °C). Each reactor was equipped with a 5.0 L gas collection bag above it, and the biogas production was measured daily using the water displacement method. The characteristics of the digestion liquid and the digestion substrate were regularly determined during the anaerobic digestion process to assess the impact of PAA on the ES and FW anaerobic co-digestion process.

2.3. Effect of PAA on each step in anaerobic fermentation

This batch experiment was conducted to investigate the impact of PAA exposure on every step of the ES and FW co-digestion process. Initially, three sets of reactors were established, each containing five fermenters of the same volume (500 mL), defined as G-I, G-II, and G-III. These sets were used to assess the effects of PAA on hydrolysis, acidification, and methanization processes. All groups were inoculated with 200 mL of inoculum and 300 mL of synthetic wastewater. PAA was then added to the reactors in varying doses to control its concentrations at 0, 3, 6, 9, and 12%. G-I's synthetic wastewater contained 2.0 g/L of bovine serum albumin (BSA) and 3.0 g/L of dextran. G-II's synthetic wastewater contained 2.0 g/L of L-alanine and 3.0 g/L of glucose. G-III's synthetic wastewater contained 2.0 g/L of sodium acetate. The impact of PAA on each anaerobic step was determined by measuring the degradation of the simulated compound in each group.

2.4. Analytical methods

The samples retrieved from the reactors were immediately passed through a 0.45-μm acetate cellulose membrane to obtain the digestion liquid for the analysis of water quality characteristics. The determination of chemical oxygen demand (COD), ammonium nitrogen (NH_4^+-N), soluble orthophosphate (SOP), TSS, and volatile suspended solids (VSS) was carried out using international standard methods (APHA 1985). Methane and VFAs were measured using gas chromatography with a chromatograph model Agilent 7890A, equipped with a flame ionization detector (FID) and a thermal conductivity detector (TCD), respectively. The calculation of methane yield is based on the amount of methane produced per unit reduction of organic matter. Specifically, it is the ratio of the cumulative methane production on a particular day to the reduction in VSS. The conditions and steps for the determination of VFA are as follows: the chromatographic column type is FFAP - MM0071, with dimensions of 30 m × 0.25 mm × 0.25 μm. The temperatures of the injection port, column oven, and detector were set at 100, 80, and 100 °C, respectively. Argon was used as the carrier gas at a flow rate of 44 mL/min, with a split ratio of 1:20 (Cheng *et al.* 2021; Kan *et al.* 2023; Li *et al.* 2023). Soluble protein (SPN) and soluble polysaccharide (SPS) were determined by the Folin phenol method and the anthrone colorimetric method, respectively, and BSA and glucose were used as standard substrates, respectively.

2.5. Statistical methods

To ensure the dependability of the results, each test was repeated three times, and the error bars were calculated from the standard deviations of these repetitions. A *p*-value less than 0.05 was set as the threshold for statistical significance, which helps to identify the differences between the sample groups.

3. RESULTS AND DISCUSSION

3.1. Effect of PAA on methane production from ES and FW anaerobic co-digestion

The changes in cumulative methane production and daily methane yield play a significant role in the operation of the reactors (Raksasat *et al.* 2021). The impact of PAA on cumulative and daily methane production is shown in Figure 1. Within each group, the cumulative methane production initially increased sharply and then stabilized. In the control group, the maximum cumulative methane production was 275 mL/g VSS, which occurred on 19 days. The addition of PAA enhanced the cumulative methane production in the co-digestion system. As the PAA content increased from 3 to 9%, the cumulative methane

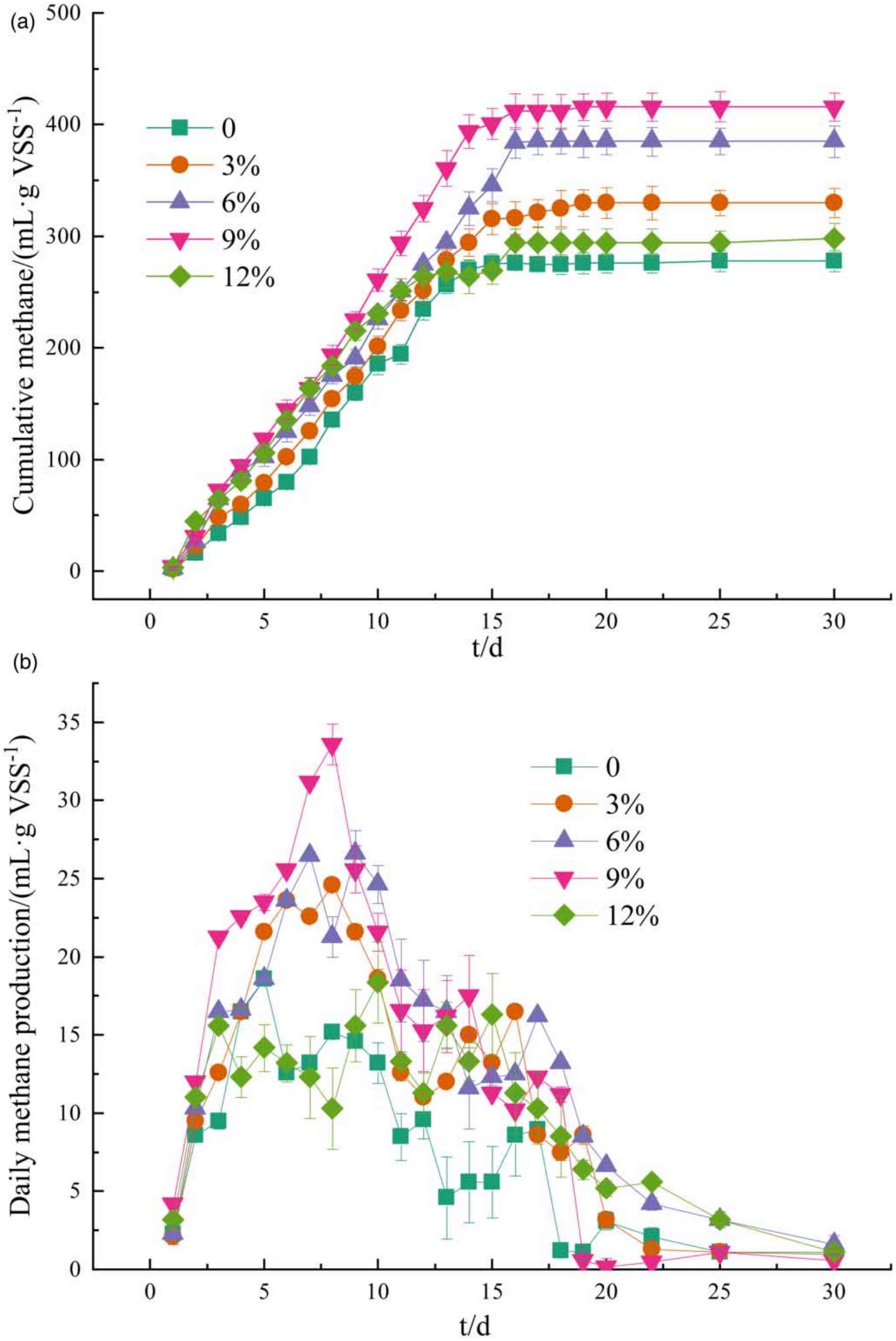

Figure 1 | Influence of PAA on the cumulative methane production (a) and daily methane yield (b) in the co-digestion system.

production rose from 320 to 416 mL/g VSS. However, when the PAA content was further increased to 12%, compared with other groups with PAA, the cumulative methane production decreased, but it remained higher than that of the control group. These experimental results confirm that PAA effectively improved the production of methane from the ES and FW co-digestion, with the optimal dose of PAA being 9%, corresponding to a methane yield of 416 mL/g VSS. Beyond 9% PAA, the co-digestion system's methane production was reduced, but it was still higher than that of the control group. Ren *et al.* (2024) applied PAA to enhance the ES anaerobic digestion, it was found that PAA altered the structure of the anaerobic digestion microbial community and increased the production of biogas. The oxidation properties of PAA can effectively accelerate the cracking of organic compounds and thus accelerate the hydrolysis process and increase the yield of methanation.

Figure 1(b) further illustrates the effect of PAA on the daily methane production during the co-digestion process of ES and FW. It can be observed that the daily methane production exhibits two peaks over time, approximately at 7 and 15 days. The first peak in daily gas production is primarily associated with the ample substrate, allowing methanogenic archaea to efficiently consume the substrate. The second peak emerges as the methanogens gradually adapt to the metabolic environment. PAA effectively enhances the daily methane production during both peaks. For instance, in the presence of PAA, the highest daily methane production increased to a range of 22.6 to 33.8 mL/g VSS, which is higher than the 18.6 mL/g VSS observed in the control group without PAA. Similar results were found at other time intervals. These findings indicate that PAA can significantly promote the efficiency of daily methane production.

PAA possesses certain oxidative properties, which effectively facilitate the solubilization of refractory organic matter within ES and FW, accelerating the hydrolysis rate and providing a material basis for methanogenic microorganisms. By enhancing the solubility of organic substrates, PAA improves the bioavailability for microbial degradation, leading to an increased rate of methane production (Ao *et al.* 2021; Guo *et al.* 2024). This suggests that PAA not only influences the immediate metabolic processes but also contributes to the overall efficiency of the anaerobic digestion system by improving the breakdown of complex organic compounds into simpler forms that can be metabolized by the methanogens, ultimately resulting in higher methane yields.

3.2. Effect of PAA on substrate dissolution process in ES and FW anaerobic co-digestion

The solubilization process of organic matter is considered to be the rate-limiting step in anaerobic digestion, as organic matter within ES and FW is predominantly present in particulate form (Zhao *et al.* 2019; Zhang *et al.* 2023a, b). The presence of PAA can accelerate the solubilization of organic matter, as shown in Figure 2, where PAA increases the concentration of soluble chemical oxygen demand (SCOD) in the co-digestion system, with a trend that higher PAA concentrations lead to more significant increases in SCOD ($p < 0.05$). Throughout the digestion cycle, the concentration of SCOD first increases and then decreases. The rise in SCOD concentration is associated with the solubilization of organic matter, while the decrease in SCOD concentration is related to the gasification process of organic matter. During the experimental process, the maximum value of SCOD increased from 1,155 to 2,584 mg/L, accompanying the increase in PAA concentration from 0 to 12%. The aforementioned experimental results clearly demonstrate that PAA promotes the release of organic matter within the co-digestion system, providing ample material assurance for subsequent acidification and methanation processes. By enhancing the solubilization of organic matter, PAA improves the efficiency of the anaerobic digestion process, making the organic matter more accessible for microbial degradation (Wang *et al.* 2024). This, in turn, leads to a more effective conversion of organic matter into biogas, primarily composed of methane and carbon dioxide, which is the ultimate goal of anaerobic digestion technology. The use of PAA as an additive in anaerobic digestion systems, therefore, holds promise for optimizing the process and increasing biogas yields, contributing to more sustainable waste management and energy production practices.

In this work, the primary organic matter within ES and FW is composed of protein (PN) and polysaccharide (PS). The investigation also examined the effects of PAA on the concentration changes of SPN and SPS. As depicted in Figure 3, similar to the pattern observed for SCOD, the concentrations of SPN and SPS increase initially and then gradually decrease over the fermentation period. Due to the higher content of PS in FW compared with PN, the concentration of SPS is higher than that of SPN within the co-digestion system. In the control group, the maximum concentrations of SPN and SPS are 784 and 645 mg/L, respectively, with the corresponding fermentation times being 11 and 13 days. The time of the appearance of the maximum values for SPN and SPS in the co-digestion system is not consistent, which is related to the different solubilization rates of PN and PS. In the presence of PAA, the maximum concentrations of SPN and SPS increase significantly to 1,644 and 1,315 mg/L ($p < 0.05$), respectively, which are much higher than those in the control group. The oxidative nature of PAA can cleave organic matter into smaller molecular substances, thereby enhancing the concentrations of SPN and SPS

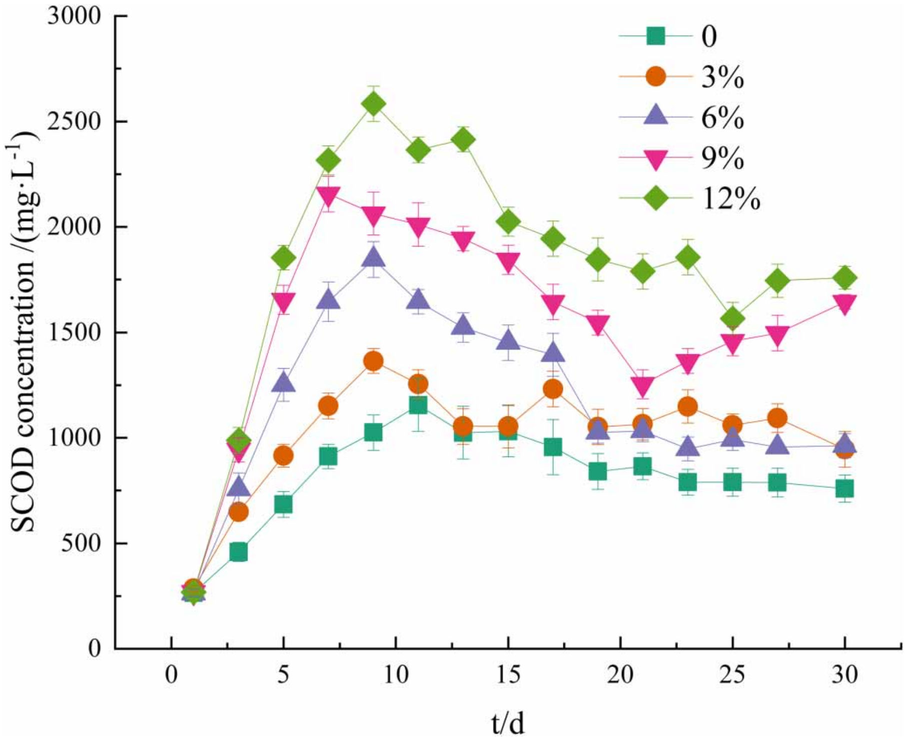

Figure 2 | Effect of AA on the concentration of SCOD in the co-digestive system of ES and FW. The error bar represents the standard deviation of the three measurements.

in the digestion liquid (Rokhina *et al.* 2010). The addition of PAA, therefore, not only accelerates the solubilization of organic matter, as evidenced by the increased SCOD, but also specifically targets the solubilization of proteins and polysaccharides, which are key components of the organic substrates in the co-digestion of ES and FW. This enhanced solubilization is crucial for improving the bioavailability of these substrates for anaerobic microorganisms, leading to more efficient degradation and conversion into methane. The findings underscore the potential of PAA as a valuable additive in anaerobic digestion processes to boost the yield and rate of biogas production.

3.3. Effects of PAA on the yield and components of VFA in the co-digestion system

VFAs are crucial intermediates in the anaerobic fermentation process of organic matter, and their production can directly affect methane yield (Li *et al.* 2019; Yuan *et al.* 2019; Qin *et al.* 2023). As shown in Figure 4, within each group, the production of VFA initially increases and then decreases over the co-digestion period. The increase in VFA is associated with the enhanced metabolic activity of acid-producing microorganisms, while the subsequent decrease in VFA is related to the metabolism of methanogenic archaea. PAA promotes the consumption of VFA, thus preventing excessive acidification. In the control group, the maximum VFA production was 2,014 mg/L, corresponding to a fermentation time of 9 days. In contrast, when PAA was present, the maximum VFA production decreased to between 1,511 and 1,974 mg/L, with higher PAA dosages leading to a more significant reduction in maximum VFA accumulation. Besides reducing the accumulation of VFA, PAA also shortened the time required for VFA to reach its maximum value, generally, the fermentation time corresponding to the maximum value of VFAs in the group with PAA presence was shortened by 1–2 days compared with the control group. It is important to note that after 20 days of co-digestion, the VFA levels in each group remained stable at 750–1,000 mg/L, indicating that the acidification stage had not been reached. The accumulation of VFA represents a dynamic equilibrium, sourced from the acidification of hydrolysis products on the one hand, and on the other hand, VFA is metabolized and volatilized by methanogenic archaea. Due to its easily degradable nature, FW is prone to excessive acidification during the anaerobic digestion process, which can lead to system collapse when severe (Li *et al.* 2019). The presence of PAA effectively reduces the accumulation of VFAs in the co-digestion system of ES and FW, thus preventing excessive acidification. In this study, the production of hydrogen gas was also detected. However, compared with the yield of methane, the yield of hydrogen gas was

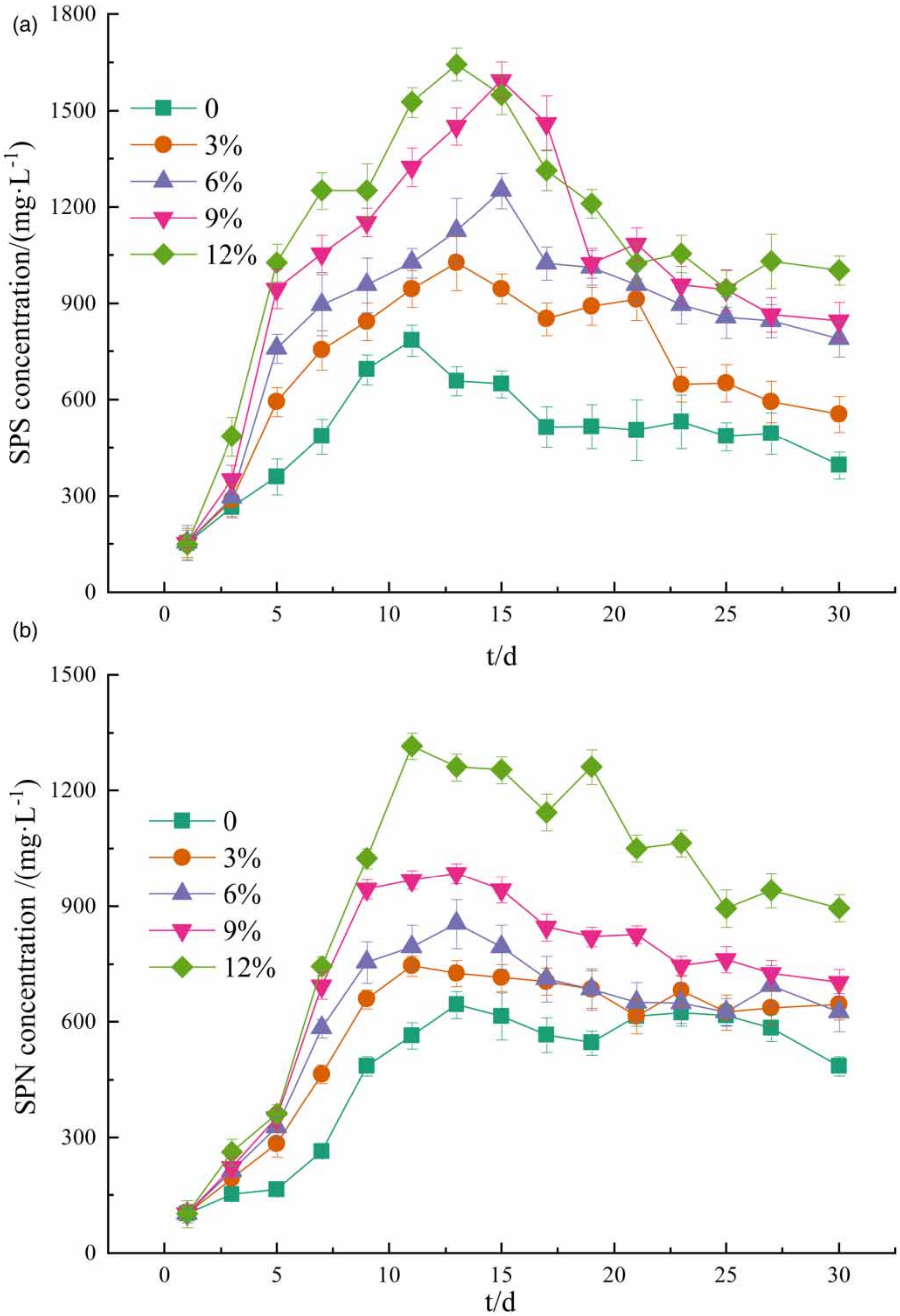

Figure 3 | Effect of PAA on the concentrations of SPS (a) and SPN (b) in the co-digestive system of ES and FW. The error bar represents the standard deviation of the three measurements.

significantly lower ($p < 0.05$). The hydrogen gas yield with PAA in this work was approximately 15.6 mL/g VSS, and the addition of PAA also enhanced the production of hydrogen.

VFAs are primarily composed of carboxylic acids with chain lengths ranging from C2 to C5. This study also investigated the impact of PAA on the proportion of individual VFA components, as shown in Figure 5. The presence of PAA increased the proportion of small molecular weight carboxylic acids. During the early stages of fermentation, a high proportion of butyrate was observed across all groups, indicating that butyric acid-type fermentation is the main fermentation type in co-digestion. PAA can influence the proportion of carboxylate salts. At 10 days, the proportion of acetate in the control group was 25.6%, while in the PAA-treated groups, the proportion of acetate ranged from 31.3 to 40.3%, which is higher than the control group.

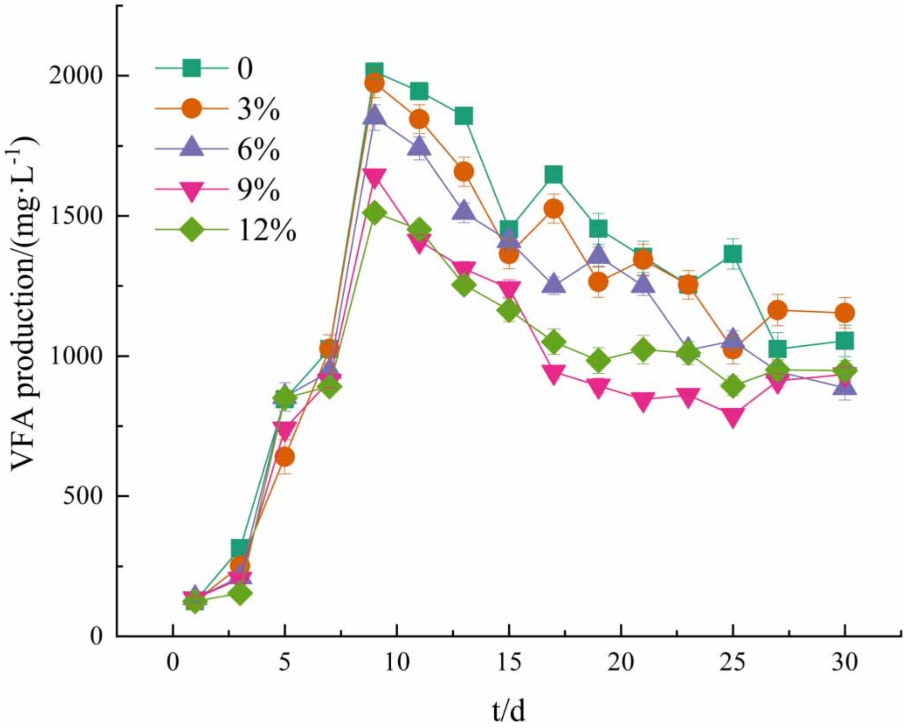

Figure 4 | Effect of PAA on the concentration of VFA in the co-digestive system of ES and FW. The error bar represents the standard deviation of the three measurements.

In the control group, the proportion of valerate, including isovalerate and *n*-valerate, was 23.3%, while in the PAA-treated groups, the proportion of valerate decreased to 14.3–20.1%, with the most significant decrease observed in the group with 9% PAA. The influence of PAA on the proportion of propionate in the co-digestion system was not significant ($p > 0.05$), as the proportion of propionate remained relatively stable at 21.3–25.8% throughout the entire process. It was found that in anaerobic digestion systems, methane is primarily produced by acetoclastic methanogens and hydrogenotrophic methanogens, with their respective contributions accounting for approximately 70 and 30% (Zhang *et al.* 2014; Ren *et al.* 2018). The increase in acetate proportion within the co-digestion system due to PAA provides a material basis for acetoclastic methanogens, which is one of the reasons why PAA enhances methane production in co-digestion. By increasing the proportion of acetate, PAA supports the activity of acetoclastic methanogens, thereby contributing to the overall efficiency of methane production in the co-digestion process.

3.4. Effect of PAA on each step of anaerobic digestion

To clarify the reasons behind the enhancement of methane production by PAA, this study also evaluated the impact of PAA on each step of the anaerobic co-digestion process. During the hydrolysis stage, BSA and dextran are degraded by hydrolytic bacteria into small molecular amino acids and glucose. As shown in Table 2, PAA increased the degradation of BSA and dextran. When the PAA concentration was 9%, the degradation efficiencies of BSA and dextran were 73.2 and 85.6%, respectively, significantly higher than the 62.2 and 72.3% in the control group. This indicates that PAA improved the hydrolysis process of organic matter, which is consistent with the results in Figure 2 where the SCOD levels in the PAA-treated groups were higher than those in the control group.

During the acidification process, various amino acids and glucose are further degraded into small molecular carboxylic acids such as acetic acid. As depicted in Figure 5(b), PAA also enhanced the degradation of L-alanine and dextran, especially in the 9% group, where the degradation efficiencies of L-alanine and dextran were 65.5 and 59.6%, respectively, again higher than the 52.3 and 49.6% in the control group. This also suggests that PAA improved the acidification metabolic process of organic matter. It should be noted that in the 12% PAA group, the degradation efficiency of the simulated substrate in the

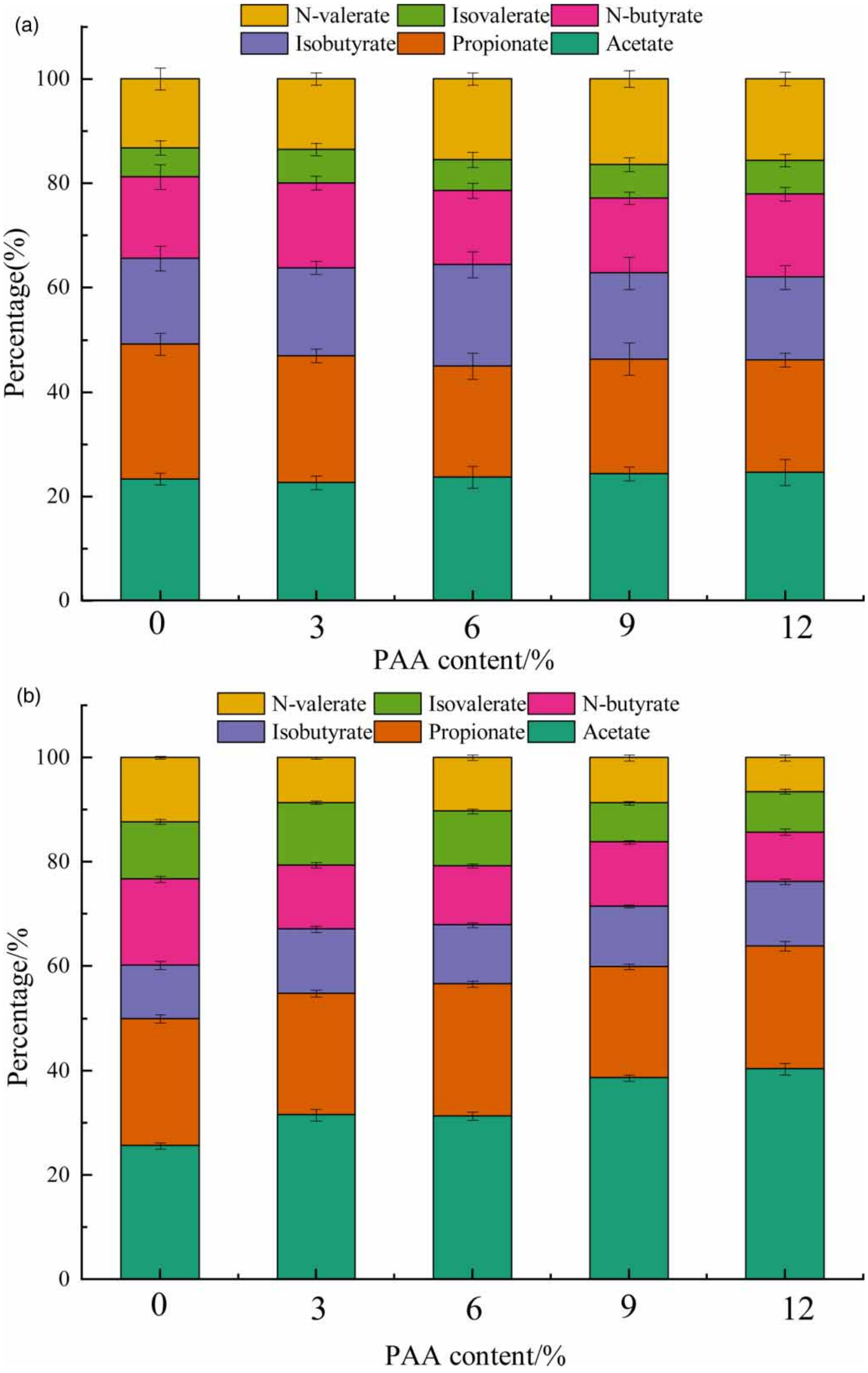

Figure 5 | Effect of PAA on the proportion of individual components of VFA in the co-digestive system of ES and FW (a: 5 days; b: 15 days). The error bar represents the standard deviation of the three measurements.

acidification process was lower than that in the group with PAA of 3–9%, but higher than that in the control group, which was related to the strong oxidizing properties of PAA, which in turn inhibited the activity of acidified microorganisms.

The promotion of hydrolysis and acidification of organic matter by PAA is related to its oxidizing properties. PAA increases the contact points between organic matter and key enzymes, enlarges the reaction area, and enhances the degradation of organic matter. The oxidizing nature of PAA not only aids in breaking down complex organic compounds into simpler

Table 2 | Effect of PAA on the degradation of simulated compounds in each step

	Degradation efficiency (%)				
Simulated substrate	**Control**	**R1**	**R2**	**R3**	**R4**
BSA	62.2 ± 2.1	68.6 ± 1.5	72.6 ± 2.6	73.2 ± 1.9	73.9 ± 2.8
Dextran	72.3 ± 3.2	78.6 ± 2.4	83.5 ± 2.1	85.6 ± 4.3	88.5 ± 4.2
L-Alanine	52.3 ± 2.8	61.3 ± 3.5	62.5 ± 2.1	65.5 ± 1.9	60.8 ± 1.8
Glucose	49.6 ± 2.8	52.3 ± 3.9	53.9 ± 2.3	59.6 ± 2.8	50.3 ± 2.2
Sodium acetate	75.6 ± 3.6	70.3 ± 4.2	62.3 ± 2.8	51.2 ± 3.2	42.6 ± 1.1

Note: The data in the table represent the average and standard deviation of the three measurements.

forms that are more accessible for microbial attack but also potentially alters the microbial community structure and function to favor methanogenic processes (Ren *et al.* 2024).

Regarding the methanogenesis process, it was found that PAA severely inhibits the methanization process, with a trend that the higher the concentration of PAA, the more pronounced the inhibition of methanization. In the group treated with 12% PAA, the degradation efficiency of acetate dropped to 42.6%, which is only 56.6% of that in the control group, indicating that PAA significantly inhibits the consumption of acetate by methanogenic archaea to produce methane. One might question why, despite the inhibition of methanogenesis by PAA, the group treated with 9% PAA showed the highest methane production. This paradox can be explained by considering the dual role of PAA in the anaerobic digestion process. While high concentrations of PAA can inhibit methanogenesis, PAA also promotes the hydrolysis and acidification processes, which provide ample substrates for methanogenesis.

In essence, the enhancement of hydrolysis and acidification by PAA leads to increased availability of VFA, particularly acetate, which is the primary substrate for acetoclastic methanogens. Even though PAA at higher concentrations can inhibit the methanogenic process, the overall effect at optimal concentrations (such as 9% in this case) may still result in higher methane production due to the increased supply of substrates. This suggests that the balance between the stimulation of earlier stages of anaerobic digestion (hydrolysis and acidification) and the potential inhibition of the methanogenic stage is crucial for optimizing methane production in the presence of PAA.

Therefore, the key to maximizing methane yield in systems employing PAA may lie in carefully controlling the concentration of PAA to ensure that the benefits of enhanced hydrolysis and acidification outweigh the inhibitory effects on methanogenesis. This finding underscores the complexity of the interactions between PAA and the microbial processes in anaerobic digestion and highlights the importance of optimizing PAA dosage for effective biogas enhancement.

3.5. The significance and inspiration of PAA to ES and FW co-digestion

Given the strong oxidizing nature of PAA, it has been utilized to enhance the oxidation and removal of refractory organic substances in wastewater, as well as in the anaerobic fermentation of waste activated sludge to produce VFA and hydrogen (Liu *et al.* 2020; Ren *et al.* 2024). In addition to these advantages, the current work further applies PAA in the anaerobic co-digestion of ES and FW to obtain the energy substrate methane, and the related mechanisms have been systematically revealed. This provides a theoretical basis for the development of treatment technologies for urban organic solid waste based on PAA processing. As far as we know, this is the first application of PAA in the ES and FW anaerobic co-digestion, and the mechanisms involved have been unveiled. This work not only provides an alternative technology for the treatment of urban organic solid waste but also further expands the application field of PAA. The application of PAA in this work showcases its versatility and potential as an environmentally friendly and efficient agent for enhancing anaerobic digestion processes. By improving the conversion of organic matter into valuable end products such as methane, PAA contributes to the circular economy and sustainable waste management strategies. The findings of this study are significant as they pave the way for the integration of PAA into existing and new waste-to-energy systems, potentially leading to more efficient energy recovery from organic waste streams and contributing to the reduction of greenhouse gas emissions.

The application of PAA to enhance the anaerobic co-digestion of ES and FW for methane production is a green and environmentally friendly strategy, as the degradation products of PAA are carbon dioxide and water, which are less harmful to the environment. PAA is capable of generating a higher amount of oxidative free radicals, which are essential precursors for the oxidation of organic matter in the sludge and FW, leading to improved solubilization and biogas production. Furthermore, various materials, including transition metals and activated carbon, can enhance the activity of PAA, thereby increasing its oxidation efficiency and accelerating the breakdown of sludge. These materials can act as catalysts, promoting the generation of more reactive oxygen species (ROS) and other free radicals that are more effective in degrading complex organic compounds. In future research, it would be beneficial to explore the development of suitable catalysts to strengthen the oxidative properties of PAA. By doing so, the efficiency of the anaerobic co-digestion process could be further improved, leading to increased methane yields and more effective treatment of organic waste. The use of catalysts could also potentially reduce the required amount of PAA, thereby lowering the operational costs and minimizing the environmental impact of the process.

Overall, the integration of PAA with appropriate catalysts presents a promising avenue for the advancement of anaerobic co-digestion technology, contributing to the sustainable management of organic waste and the production of renewable energy in the form of biogas.

4. CONCLUSION

This work reports a new strategy for enhancing methane production from the anaerobic co-digestion of ES and FW using PAA, and reveals the related mechanisms. The optimal dose of PAA is 9%, with the maximum methane yield being 416 mL/g. The methane production in the PAA-treated group is higher than that of the control group, and the fermentation time is also shortened to varying degrees. PAA promotes the release of organic matter within the co-digestion substrate and increases the concentrations of SPN and SPS. PAA facilitates the production and conversion of VFA, preventing excessive acidification, thereby increasing methane yield. Additionally, PAA promotes substrate degradation in hydrolysis and acidification process, but reduces substrate consumption in the methanogenic process. PAA is proven to be a green and efficient strategy for promoting methane production from the co-digestion of ES and FW.

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AUTHOR CONTRIBUTIONS

X.Y.: Writing – original draft. J.F.: Writing – review and editing. Y.X.: Writing – review and editing, Project administration.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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