

MEC-AD 利用衍生碳高效产甲烷：电极、促进剂与 产甲烷途径的新见解

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摘 要: 在微生物电解池耦合厌氧发酵系统(MEC-AD)中, 以钛网为电极, 探究不同电压(0、0.4、0.6、0.8、1.2 V)对系统性能的影响, 发现 0.8 V 电压下沼气产量与有机物降解率最佳。合成芦荟废弃物衍生的球形碳(AVW-SC)与多孔碳(AVW-PC), 分别涂覆于钛网作为 MEC-AD 电极。结果表明, 以涂覆 AVW-PC 钛网做电极的 MEC-AD 系统发酵性能更优。研究不同涂覆量(0.025、0.05、0.10、0.15、0.20 g)AVW-PC 对 MEC-AD 系统的影响, 当 AVW-PC 涂覆量为 0.10 g 时, MEC-AD 系统($Ti_{0.8}-PC_{0.1}$)的累积沼气产量与甲烷含量均达到最高。在 $Ti_{0.8}-PC_{0.1}$ 中添加不同剂量的 AVW-PC(0.10%、0.15%、0.20%、0.25%, 质量分数)作为促进剂, 当 AVW-PC 的添加量为 0.20%时, MEC-AD 系统($Ti_{0.8}-PC_{0.1}/PC_{0.2}$)在产气量(633.63 mL/g VS)、甲烷含量(65.85%)及沼渣总营养物含量($42.30\text{ g}\cdot\text{kg}^{-1}$)方面表现最优。 $Ti_{0.8}-PC_{0.1}/PC_{0.2}$ 中 *Bacteroidales*、*Pseudomonadales*、*Oscillospirales*、*Methanobacteraceae*、*Methanospirillaceae*、*Methanosarcinaceae* 和 *Methanosaetaceae* 的丰度显著提高。微生物多样性的增加促进了种间氢转移、种间乙酸转移和直接种间电子转移, 进而提高了产甲烷效能, 揭示了 AVW-PC 作为电极与促进剂对甲烷增产的双重作用机制, 为生物质衍生碳在 MEC-AD 系统中的多功能应用提供了新思路。

关键词: 芦荟废弃物; 生物质衍生碳; 电极; 促进剂; 甲烷生产

中图分类号: O611.62 文献标志码: A 文章编号: 0454-5648(2025)12-3740-21

网络出版时间: 2025-11-06



1 Introduction

The production of biological waste, such as agricultural waste and animal manure, has resulted in serious environmental pollution. In China, the overabundance of animal waste, especially cattle dung (CD), adversely affects the environment. Animal compost contains various microorganisms, hormones, and harmful substances that can contaminate water sources^[1]. Meanwhile, extensive aloe vera cultivation in China supplies gel products for multiple sectors but it also yields a large amount of aloe vera waste (AVW)^[2]. Therefore, the rational utilization of this waste is crucial for alleviating environmental problems and reducing resource waste^[3]. Owing to their high biodegradability, these organic wastes can act as co-substrates in anaerobic co-digestion (AcoD) to produce clean energy that can replace fossil fuels^[4]. Several researchers have

demonstrated that AcoD shows more favorable performance than monodigestion with regard to gas production and the extent of biodegradation within the anaerobic digestion environment^[5]. In most cases, co-digestion significantly enhances biogas production compared with digestion with a sole substrate because of constructive interactions within the digestion environment and essential nutrient provision^[6]. Co-digestion has advantages but still faces challenges such as low methane production, instability, and long digestion times, which require further exploration and improvement.

There has been noteworthy progress regarding a system that integrates MEC-AD to enhance biogas production via the capture and conversion of carbon dioxide gas into methane gas^[7]. Microbial electrolytic cells (MEC) enrich the electroactive microorganisms on both electrodes and in the native solution, thereby facilitating CH_4 formation. Electroactive microorganisms

收稿日期: 2025-02-19。 修订日期: 2025-03-21。

基金项目: 国家重点研发计划(No.2018YFB1502900); 陕西省重点研发计划国际合作重点项目(No.2019KWZ-03); 陕西省重点科技创新团队(No.2022TD-34); 青海省高原绿色建筑与生态社区重点实验室开放基金项目(No. KLKF-2019-002)。

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Received date: 2025-02-19. Revised date: 2025-03-21.

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expedite electron transfer in the reaction environment, thereby fostering biofilm formation and creating a conducive methanogenic environment^[8–9]. Electrode materials are integral to anaerobic digestion (AD) performance efficacy. Zheng et al.^[10] revealed that the characteristics of cathode materials significantly impact biofilm adhesion. An improved bioelectrochemical process was achieved using a bimetallic hybrid cathode (Ni/Co-NC) with a carbon foundation. This cathode accelerates hydrogenotrophic methanogenesis, allowing for direct electron transfer. A comparative analysis of the performance characteristics of distinct biological cathode-carbon brushes and graphite plates found a notably higher CH₄ production rate with a carbon brush compared to a reactor equipped with a graphite electrode. Cathodes composed of carbon materials exhibit exceptional electrochemical characteristics and superior microbial adsorption capacity, surpassing metal cathodes for CH₄ generation^[11].

The addition of accelerants is another method for optimizing the efficiency of the MEC-AD system. Carbon derived from biomass has been extensively utilized in various dye-sensitized solar cells^[2], supercapacitors^[12], and AD reactors^[13]. Given its economic viability and widespread availability, bio-based carbon is an ideal choice as an accelerant in MEC-AD systems. Bio-based carbon facilitates electron transfer pathways among microorganisms, thereby enhancing digestion performance^[14]. Additionally, these materials provide a conducive growth environment for microorganisms, enriching their populations and promoting substrate degradation, thereby improving the digestion efficiency^[2,7]. Furthermore, bio-based carbon materials demonstrate robust capabilities in fostering interactions among anaerobic microorganisms, facilitating enhanced electron transfer rates^[15–16]. Meanwhile, some carbon materials adsorb CO₂ in biogas for upgrading purposes. This is due to the pore and surface properties of activated carbon, enabling the physical adsorption of CO₂ in micropores, reducing CO₂ content, and increasing methane purity. There are some reports on carbon materials such as electrodes or accelerants. However, the effect of biomass-derived carbon serving as both an accelerant and an electrode on the performance of MEC-AD is not yet known, and the synergistic effect of bio-based carbon electrodes and accelerants on electron transfer and methanogenic pathways remains to be revealed. Thus, employing bio-based carbon as both electrodes and accelerants in MEC-AD systems presents a viable approach for biogas

upgrading and realizes multifunctional utilization, which is worth studying.

This study investigates how biomass-derived carbon serves as both an accelerant and an electrode that influences the performance of MEC-AD. This explains how bio-based carbon improves biogas and methane production through interspecies hydrogen transfer (IHT), interspecies acetic transfer (IAT), and direct interspecies electron transfer (DIET). Both spherical carbon (SC) and porous carbon (PC) were produced from AVW and studied using scanning electron microscopy (SEM), X-ray diffraction (XRD), and Raman spectroscopy. AVW-SC and AVW-PC were then employed as electrodes and accelerants, respectively, and their effects were evaluated through biogas production assessment, total chemical oxygen demand (TCOD) removal, removal rate of total solids (TS) and volatile solids (VS), and pH variation. The fertility and stability of the digestate were evaluated by measuring total nitrogen, phosphorus, and potassium. Further investigations were conducted on the functions of accelerants and bio-based carbon electrodes in the electron transfer and methanogenic pathways through electrochemical testing and microbial community investigations. Finally, the synergistic effect of bio-based carbon acting as both an electrode and an accelerant in the MEC-AD system was elucidated.

2 Materials and methods

2.1 Co-substratum and inoculum

AVW and CD were used as co-substrates in this work. The CD, taken from a sizable dairy breeding center in Xi'an, China, was wrapped in black bags and kept at room temperature for a week to adapt the anaerobic environment^[17]. AVW was sourced from aloe vera cultivation in Yangling, China. NaOH pretreatment has been proven to decompose the ester bonds of cellulose, hemicellulose, and lignin for easy degradation based on the optimized conditions of alkaline pretreatment indicated by Yun et al.^[18–19] AVW samples were crushed into small particles approximately 5 mm in diameter. These particles were steeped in a 2 mol/L NaOH solution for 24 h and then were washed with distilled water to remove alkali residues and achieved the pH of 7 before being mixed with cattle dung. The sewage sludge utilized as inoculum was gathered from a wastewater treatment facility in Xi'an, China. Prior to the experiments, all the feedstocks were homogenized and stored at room temperature. Table 1 shows comprehensive characterization of both the co-substratum and inoculum.

Table 1 Characteristics of cattle dung, aloe vera waste, and sewage sludge

Parameters	w (TS)/%	w (VS)/%	w (VS/TS)/%	pH	w (Carbon)/%	w (Hydrogen)/%	w (Nitrogen)/%	w (C/N)/%
Cattle dung	15.29±0.61	11.50±0.09	75.33±0.35	7.86±0.04	47.28±0.12	5.72±0.10	4.20±0.20	11.26±0.60
Aloe vera waste	12.17±0.40	9.72±0.26	79.90±0.50	7.03±0.08	59.20±0.33	11.40±0.15	1.52±0.03	38.95±0.50
Sewage sludge	4.29±0.12	3.51±0.14	81.90±0.53	7.16±0.07				

w—Mass fraction.

2.2 Catalyst preparation and electrode fabrication

2.2.1 Preparation of bio-based carbons AVW–SC and AVW–PC were synthesized by hydrothermal method with AVW as raw material (Fig. 1). Firstly, AVW was washed, dried and ground to powder. Mixed with DI water at 1 : 20, the AVW powder was stirred at 25 °C for 4 h, then placed in a high-pressure reactor and heated hydrothermally at 200 °C for 5 h to get a solid–liquid mixture. After filtration, washing with ethanol and DI water and drying, AVW–SC was obtained. For AVW–PC, AVW–SC was uniformly mixed with KOH and deionized water in a mass ratio of 1 : 2 : 20. The blend was stirred for 4 h and dried. The dried powder was put in a quartz container and pyrolyzed in a tube furnace (800 °C, 2 h). The pyrolyzed powder was washed with diluted

hydrochloric acid and pure water to neutralize and dried at 105 °C to yield AVW–PC.

2.2.2 Electrode fabrication The simplified preparation procedure (Fig. 1) for spherical and porous carbon electrodes is as follows. First, the prepared bio-based carbon (either spherical or porous) is mixed with 5% polytetrafluoroethylene powder in appropriate proportions in ethanol and deionized water to form a mixed solution. Subsequently, the mixture is subjected to ball milling for 4 h to obtain an ink-like slurry. The slurry was repeatedly and evenly sprayed onto a 2 cm × 3.5 cm Ti mesh to get a coated mesh. After 24 h of oven drying, the electrode was sintered at 360 °C for 1 h in N₂ to prevent coating shedding. Finally, the spherical carbon electrode (Ti–SC) and porous carbon electrode (Ti–PC) were obtained.

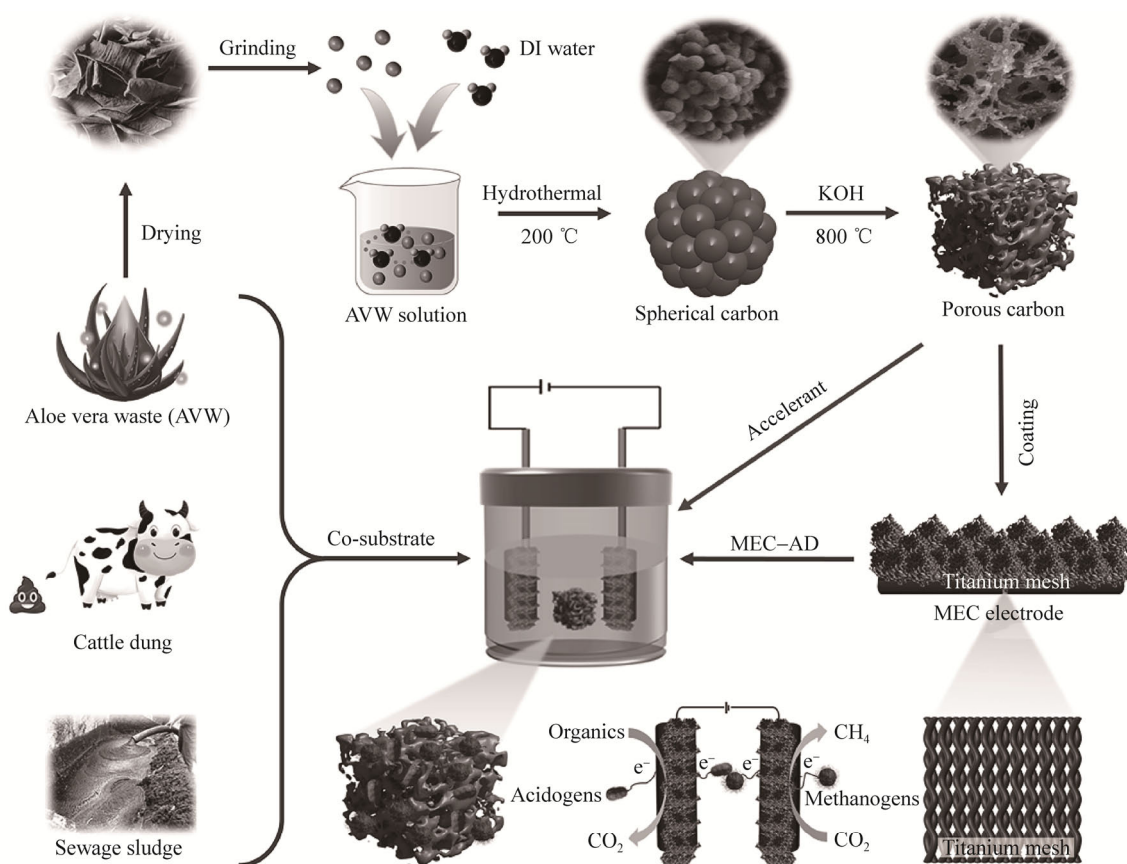


Fig. 1 Schematic illustration of the strategy for preparing AVW–SC and AVW–PC, which were coated Ti mesh as electrode and added into the AD pool as the accelerants in MEC–AD system

2.3 Experiments set up for MEC–AD

As depicted in Fig. 1, a 500 mL wide-mouth glass bottle with a 400 mL working volume functioned as the MEC–AD reactor. Two fabricated electrodes, Ti–SC and Ti–PC, spaced 2 cm horizontally, were inserted into the reactor through a rubber plug to act as the anode and cathode respectively. Each MEC–AD system was filled with a co-substrate mixture of cattle dung and aloe vera waste and inoculum of sewage sludge in a 3:7 mass ratio, then placed in a thermostatic water bath at (36±1) °C for 35 d. For biogas measurement, a rubber hose connected

each system to a conical flask filled with water, allowing the biogas to displace water in the flask. The electrodes were wired to an external 0–1.2 V DC power supply using crocodile clips and titanium wire (TA1, $\phi=3$ mm), and a digital multimeter was plugged into the electrodes to monitor the current and ensure stable voltage supply.

In this work, four batches of experiments were conducted. First, to find the optimal voltage, Ti meshes (labeled Ti₀, Ti_{0.4}, Ti_{0.6}, Ti_{0.8}, Ti_{1.2}) were used as electrodes under 0–1.2 V. Second, at the optimal voltage, Ti meshes coated with 0.05 g of AVW–SC or AVW–PC

were tested to select the best carbon electrode. Third, varying coating amounts (0.025–0.200 g) of the chosen material were examined at the optimal voltage to determine the ideal coating quantity. Finally, AVW–SC or AVW–PC accelerants (0.10%–0.25%) were added. Systems with coated electrodes at the optimum voltage were named Ti_X-SC_Y and Ti_X-PC_Y , after adding accelerant, they were labeled Ti_X-PC_Y/PC_M or Ti_X-SC_Y/SC_M . X is the optimum voltage, Y the optimal coating amount, and M the accelerant concentration. Carbon rods as electrodes were the control group (CK_{CR}). All experiments were repeated three times.

2.4 Analysis methods

2.4.1 Characterization and electrochemical assessments
The X-ray powder diffractometer (D/MAX–2200, MDI, USA) was employed to test the XRD patterns of AVW–SC and AVW–PC. The morphological features of AVW–SC and AVW–PC were examined by SEM. Using a standard three-electrode setup, tests such as cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were performed on the manufactured electrodes. The isotherms were obtained by the Barrett–Joyner–Halenda (BJH) method to derive the pore size distributions, and the specific surface areas were calculated by the Brunauer–Emmett–Teller (BET) method.

The following formulas are used to calculate the electron-donating capacity (EDC) and the electron-accepting capacity (EAC), which obtained the current-time ($I-t$) curve in the electron collection process of the tested sample through the electrochemical workstation, and the number of electrons obtained by integrating the $I-t$ curve represents the ability to accept electrons (EAC) and donate electrons (EDC) of AVW–SC and AVW–PC^[2, 7, 20].

$$EAC = \frac{\int \frac{I_{red}}{F} dt}{m} \quad (1)$$

$$EDC = \frac{\int \frac{I_{ox}}{F} dt}{m} \quad (2)$$

After adjusting the initial baseline, the reduction current in MER is I_{red} , while the oxidation current in MEO is I_{ox} . The mass of AVW–PC and AVW–SC is represented by m , and the Faraday constant is F (96 485 C/mol).

2.4.2 Sampling and chemical methods Biogas production was quantified and calculated using the drainage gas collection method. The biogas composition was examined by a gas chromatograph (GC9790, China). The pH was determined by a portable, high-precision pH acidimeter (Ph–02SL, China). TS, VS, and TCOD were quantified following established protocols (APHA, 2005). The thermogravimetric (TG), differential scanning calorimetry (DSC) and differential thermogravimetric analysis (DTG) were examined using a differential thermal analyzer (STA449F3, Germany). The total

nitrogen (TN), total phosphorus (TP), and total potassium (TK) contents of airdried digestates were measured by an automatic Kjeldahl apparatus (JK9830, China), a spectrophotometer (Ruili VIS-723 N, China) and an atomic absorption spectrometer (AA-7003, China), respectively.

2.4.3 Microbial community analysis To elucidate the characteristic alterations in microbial communities, high-throughput testing was used to analyze the samples. DNA was extracted using universal 16S rRNA gene primers: forward (CCTACGGGNGGCWGCAG) and reverse (GACTACHVGGGTATCTAATCC). The sequence of data mentioned has been stored in SRA under the code 16S237664XA.

2.5 Statistical analysis

The modified Gompertz model (Eq. (3)) was utilized to forecast the kinetic parameters obtained from the MEC–AD system.

$$P(t) = P_m \times \exp \left\{ -\exp \left[\frac{R_m \times e}{P_m} (\lambda - t) + 1 \right] \right\} \quad (3)$$

Where $P(t)$ is the cumulative biogas yield (mL/g VS) at time t . P_m is the biogas yield potential (mL/g VS). R_m is the maximum biogas yield rate (mL/g VS/d). λ is the lag phase. t is the digestion time (d), and e is a constant (2.718 28).

The coulombic efficiency (CE) is calculated by equation (4)^[21]. The total methane recovery (R_{tot}) is based on substrate consumption to the maximum theoretical yield, calculated by equation (5)^[22].

$$CE(\%) = \frac{M \int_0^t I dt}{n \cdot F \cdot V \cdot \Delta COD} \times 100\% \quad (4)$$

$$R_{tot}(\%) = \frac{V_{CH_4}}{0.35V \Delta COD} \quad (5)$$

where M represents the molar mass of acetate (60 g/mol). I is the current (A), which was calculated according to the daily measured value. t is time (d). n denotes the number of moles of electrons needed to oxidize 1 mol of COD. F is the Faraday constant (96 485 C/mol). V is the working volume of the reactor (L) and ΔCOD is the change in COD during the experiment (g/L). V_{CH_4} represents the volume of methane generated.

3 Results and discussion

3.1 Properties of AVW–SC and AVW–PC

The SEM image of AVW–SC in Fig. 2(a) shows that the AVW–SC is spherical and tightly aggregated, and the particle size is about 300 nm–1 μ m. AVW–PC shows a three-dimensional networked structure and exhibits a larger specific surface area compared to spherical structure AVW–SC in Fig. 2(b), which is conducive to energy collection, conversion and storage, thereby improving electron exchange capacity. In XRD pattern in Fig. 2(c), the synthesized AVW–SC has an obvious diffraction peak near $2\theta=24.3^\circ$, which is caused by the

(002) lattice spacing of amorphous carbon. In addition, organic acids, like CaC_2O_4 were also identified based on additional diffraction peaks, which were derived from biogenic organic acids contained in aloe vera. The two

broad peaks near 24.3° and 43.3° in the XRD pattern of AVW-PC presented in Fig. 2(d). The XRD and SEM analysis confirmed that the aloe vera waste-derived AVW-SC and AVW-PC were successfully prepared.

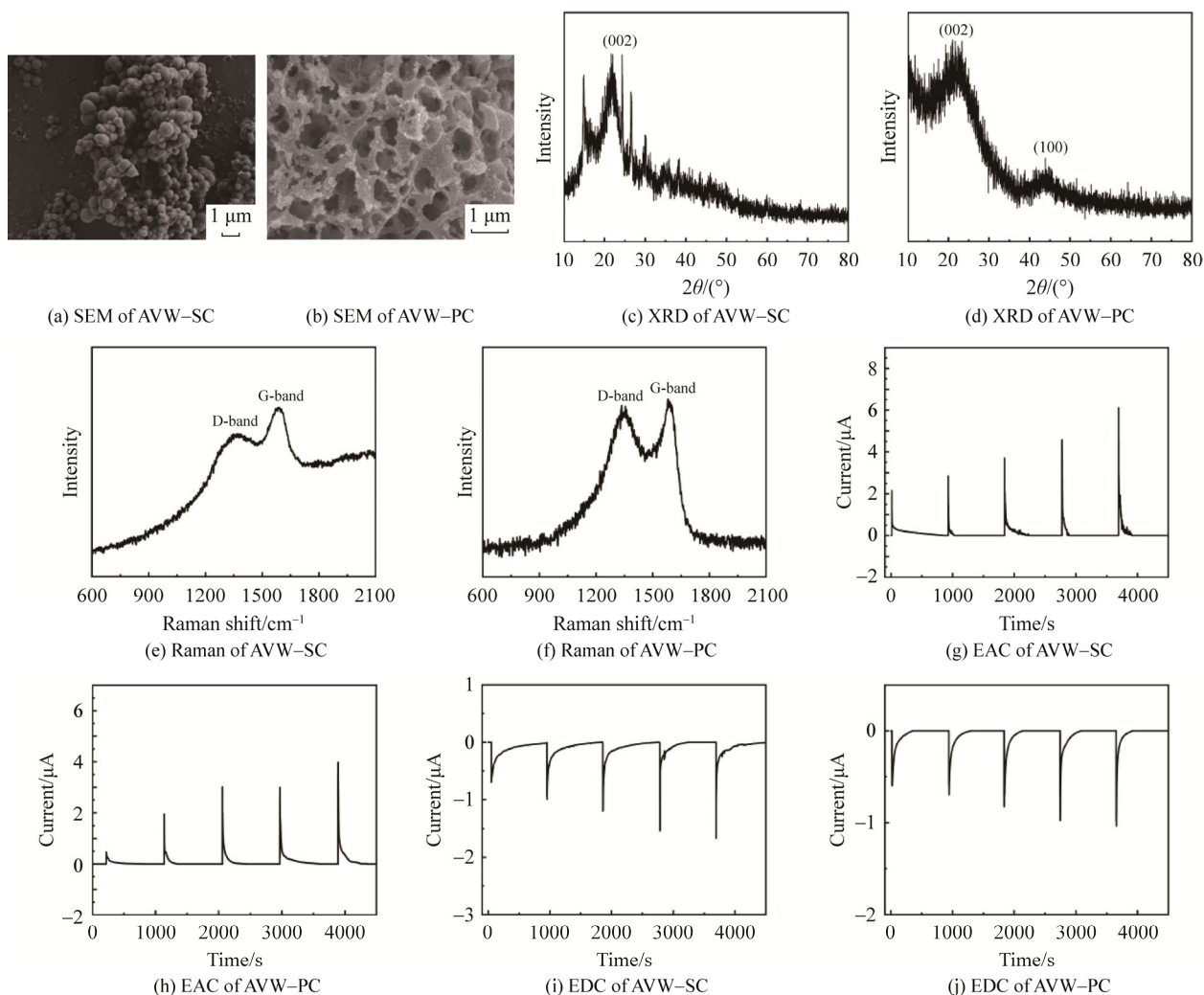


Fig. 2 Characterization of AVW-SC and AVW-PC

The Raman spectra is shown in Fig. 2(e) and Fig. 2(f). The D-band is ascribed to the respiration mode vibrations of A_{1g} . Another peak at 1580 cm^{-1} for AVW-SC and AVW-PC corresponds to the E_{2g} vibrational mode. Ratio (I_D/I_G) indicates degree of defects in material^[23]. The value of I_D/I_G for AVW-PC (0.97) exceeds that of AVW-SC (0.90), which is attributed to an increase in porosity and disruption of the sp^2 graphite structure^[2, 24]. Raman diagrams (Figs. 2(e)–2(f)) show that AVW-PC has a higher degree of graphitization than AVW-SC. A higher degree of graphitization improves the conductivity of AVW-PC, making it a better ability to exchange electrons^[2, 7].

The redox characteristics of AVW-SC and AVW-PC were assessed using mediated electrochemical oxidation (MEO) and reduction (MER). As depicted in Figs. 2(g)–2(j), the reduction (oxidation) currents mediated by peaks of AVW-SC and AVW-PC show a

significant increase with the rising concentration of accelerant. The EAC of AVW-SC and AVW-PC amounted to 0.09 and $0.31\text{ }\mu\text{mol}\cdot\text{e}^-/\text{g}$. The EDC for AVW-SC and AVW-PC was 0.06 and $0.44\text{ }\mu\text{mol}\cdot\text{e}^-/\text{g}$. Consequently, AVW-SC and AVW-PC have electron exchange capacity (EEC) of 0.15 and $0.75\text{ }\mu\text{mol}\cdot\text{e}^-/\text{g}$, which was similar to the biomass derived carbon previously reported^[25]. This result indicates that AVW-PC has better electron-donating and electron-accepting ability than AVW-SC, which is admirable for the electron transfer during microbial metabolism. Superior EEC may enable AVW-PC accelerant to accelerate DIET between acetogenic bacteria and methanogenic archaea, thereby promoting methane production.

The nitrogen adsorption–desorption isotherms of AVW-PC are shown in Figs. 3(a)–3(b). They exhibit type IV nitrogen adsorption–desorption isotherms, which are typical of mesoporous materials according to the IUPAC

classification^[26]. The BET surface area of AVW-PC is 641.80 m²/g, the adsorption average pore diameter is 9.77 nm. These values are consistent with previous research findings^[27], suggesting that the synthesized AVW-PC has a high porosity and a large specific surface area. The high BET specific surface area indicates that

AVW-PC can provide more sites for substrate accumulation and microbe enrichment, which can provide a more favorable growth environment for bacteria and archaea, thereby improving the efficiency of AD systems. This will be confirmed in the next biogas production measures.

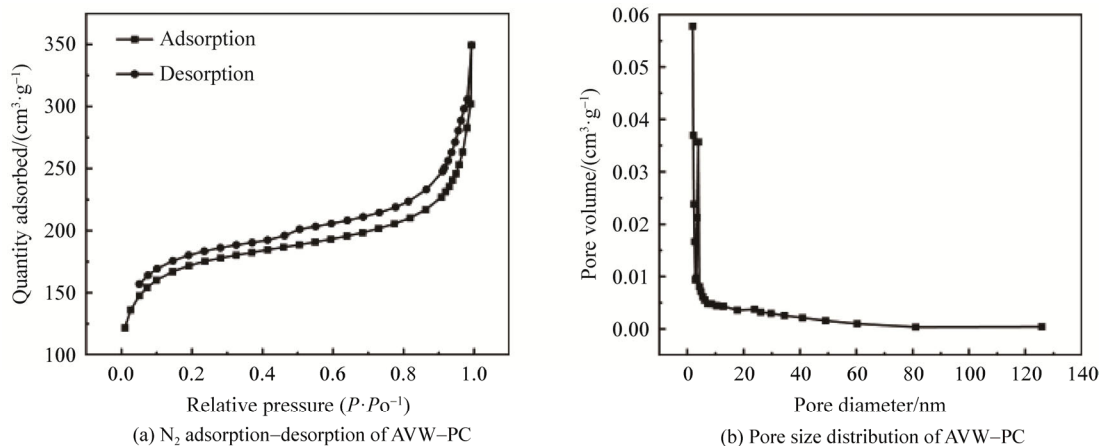


Fig. 3 Nitrogen adsorption-desorption isotherm and pore size distribution of AVW-PC at 77.30 K

3.2 Property of AVW-SC and AVW-PC coated Ti mesh as electrode on MEC-AD system

Firstly, we used Ti mesh as the electrode and applied different voltages to explore a suitable voltage for subsequent experiments. After 35 d of digestion, the daily biogas production, cumulative biogas production, pH, TCOD, TS and VS removal rates of MEC-AD system with Ti mesh electrode under various voltages are shown in Fig. 4. The daily production of biogas showed two gas production peaks in Fig. 4(a). The first peak, linked to simple organic matter degradation, emerged between 10 and 16 d; the second peak, related to complex organic matter degradation, appeared between 19 d and 24 d. Notably, when the voltage was 0.8 V, the maximum daily biogas yield of 35.44 mL/g VS was obtained on the tenth day, and the highest cumulative biogas yield of 424.01 mL/g VS was obtained in Fig. 4(b) after the entire 35-day digestion cycle.

The change trend of pH in the digestion system in Fig. 4(c) was recorded every 5 d. At the beginning of digestion, the pH was between 7.76–7.94. In the initial five days, the pH exhibited a downward trend, which was due to the accumulation of VFA caused by the hydrolysis of a substantial quantity of organic matter in the hydrolysis stage, resulting in the acidification of the environment. Then the pH increased once more as a result of the methanogens consuming VFAs. The application of 0.8 V voltage in the Ti_{0.8} group expedited the pH recovery, promoting the growth of methanogens more rapidly compared to the Ti₀, Ti_{0.4}, Ti_{0.6}, and Ti_{1.2} groups. Fig. 4(d) shows the TCOD removal rate of MEC-AD system with Ti mesh as electrode under different voltages. The TCOD removal rates are 33.27% for Ti₀, 38.05% for Ti_{0.4}, 42.62% for Ti_{0.6}, 47.62% for

Ti_{0.8} and 45.88% for Ti_{1.2} groups, respectively. When the voltage is 0.8 V, the Ti_{0.8} group exhibited a higher TCOD degradation rate compared to other groups, suggesting a more through consumption of organic matter within the Ti_{0.8} group. Higher TS removal rates (37.50%) and VS removal rates (48.55%) are obtained in Figs. 4(e)–4(f), which are higher than those of other groups, reflecting the substrate degradation more completely in the whole MEC-AD system under the applied voltage of 0.8 V.

In general, the MEC-AD system with Ti mesh as the electrode obtained the superior production of biogas and breakdown of organic material when a voltage of 0.8 V is applied. This means that an applied voltage set at 0.8 V is optimal voltage, which will be fixed in the following experiments.

On this basis, the electrochemical properties of two kinds of carbon electrodes at 0.8 V voltage were investigated. The current response of different MEC-AD systems is shown in Fig. 5(a). Stable current indicated that a mature electroactive biofilm is formed on the electrode^[10]. After 35 d, substrate consumption led to reduced methanogen, causing current gradually decrease. The coulombic efficiency (CE) was measured to evaluate microbial electron transfer efficiency^[28]. Ti_{0.8}-PC_{0.05} demonstrated a 19.17% improvement in CE compared to Ti_{0.8}-SC_{0.05}, possibly due to the AVW-PC catalyst enhancing electrode biocompatibility, thereby enhancing redox reactions with microorganisms in MEC-AD, similar to that is observed previously^[29]. This indicates higher electron diffusion rates on the AVW-PC electrode surface. Moreover, Ti_{0.8}-PC_{0.05} exhibited superior methane recovery (R_{tot}), reaching 77.60%, surpassing the methane recovery rates of 71.04% for Ti_{0.8}-SC_{0.05}, 73.49% for Ti_{0.8}, and 75.65% for CK_{CR}, as illustrated in Fig. 5(b).

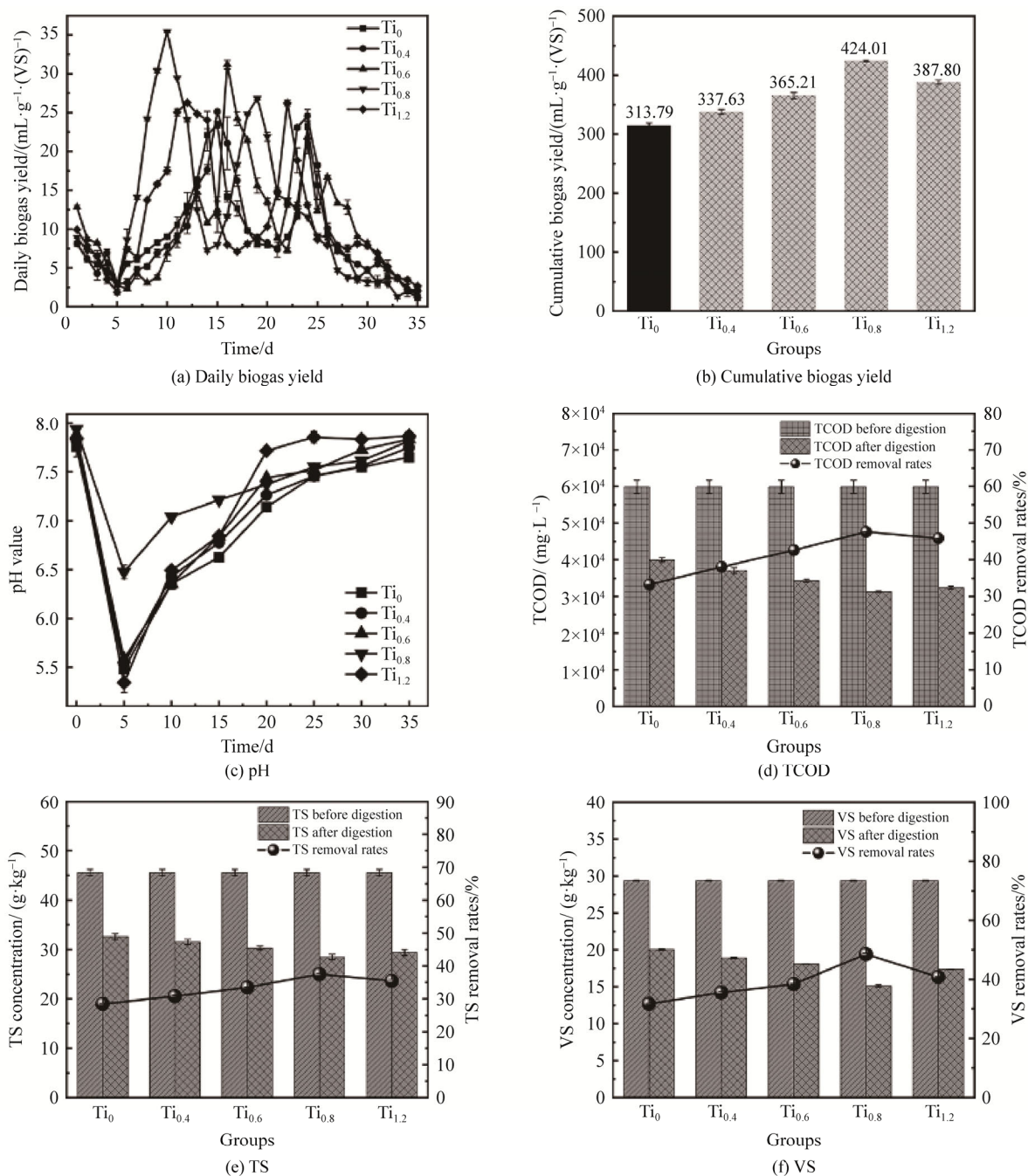


Fig. 4 Daily biogas yield, cumulative biogas yield, pH, TCOD, TS, and VS of MEC-AD system with pure Ti mesh electrode under different voltages

The electrochemical performance of CK_{CR}, Ti_{0.8}, Ti_{0.8}-SC_{0.05}, and Ti_{0.8}-PC_{0.05} were investigated with digestate as electrolyte. The CV test was applied to evaluate the interaction and capacitive properties of electron transfer in electrode biofilms^[30]. The region beneath the CV curve is correlated with the capacitance per unit area of the corresponding pole piece^[31]. The area of the CV profile increases in the order of CK_{CR} < Ti_{0.8} < Ti_{0.8}-SC_{0.05} < Ti_{0.8}-PC_{0.05} in Fig. 5(c), which proves the optimal energy storage capacity of the Ti_{0.8}-PC_{0.05}. The good capacitance performance of the Ti_{0.8}-PC_{0.05} can be attributed to the substantial pore

structure of AVW-PC, which is testified and illustrated in Fig. 2(b). However, CK_{CR} and Ti_{0.8} had no obvious redox peaks in the scanning range. The oxidation peak currents of Ti_{0.8}-SC_{0.05} and Ti_{0.8}-PC_{0.05} are 3.54 and 5.43 mA, respectively, while their reduction peak currents are -5.06 and -9.33 mA, respectively. These results indicate that Ti_{0.8}-SC_{0.05} and Ti_{0.8}-PC_{0.05} achieve excellent redox activity in MEC-AD system, which is in sharp contrast to MEC-AD system with Ti_{0.8} and CK_{CR}.

To further understand the electrochemical properties, the resistance to transfer of electric charge reactions of the biofilm on the electrode was analyzed by

EIS. Meanwhile, the Nyquist plot radius indicates internal resistance. The internal resistance of the $\text{Ti}_{0.8}\text{-PC}_{0.05}$ electrode is the smallest in Fig. 5(d), with an internal resistance value of $3.00\ \Omega$, followed by the $\text{Ti}_{0.8}\text{-SC}_{0.05}$ ($12.01\ \Omega$), $\text{Ti}_{0.8}$ ($12.64\ \Omega$), and CK_{CR} ($16.80\ \Omega$), indicating that the charge transfer resistance on the $\text{Ti}_{0.8}\text{-PC}_{0.05}$ is reduced, thereby accelerating the electron transfer of MEC-AD system. Fit the EIS Nyquist diagram using the relevant elements and then

tabulate these elements along with their respective values, as shown in Table 2.

Above all tests in Fig. 5 confirmed that the $\text{Ti}_{0.8}\text{-PC}_{0.05}$ group showed the best biogas production performance, substrate degradation, coulomb efficiency, methane recovery, and the electrochemical properties as compared with other groups, the $\text{Ti}_{0.8}\text{-PC}_{0.05}$ is a more suitable electrode material than other groups (CK_{CR} , $\text{Ti}_{0.8}$, and $\text{Ti}_{0.8}\text{-SC}_{0.05}$) for the MEC-AD.

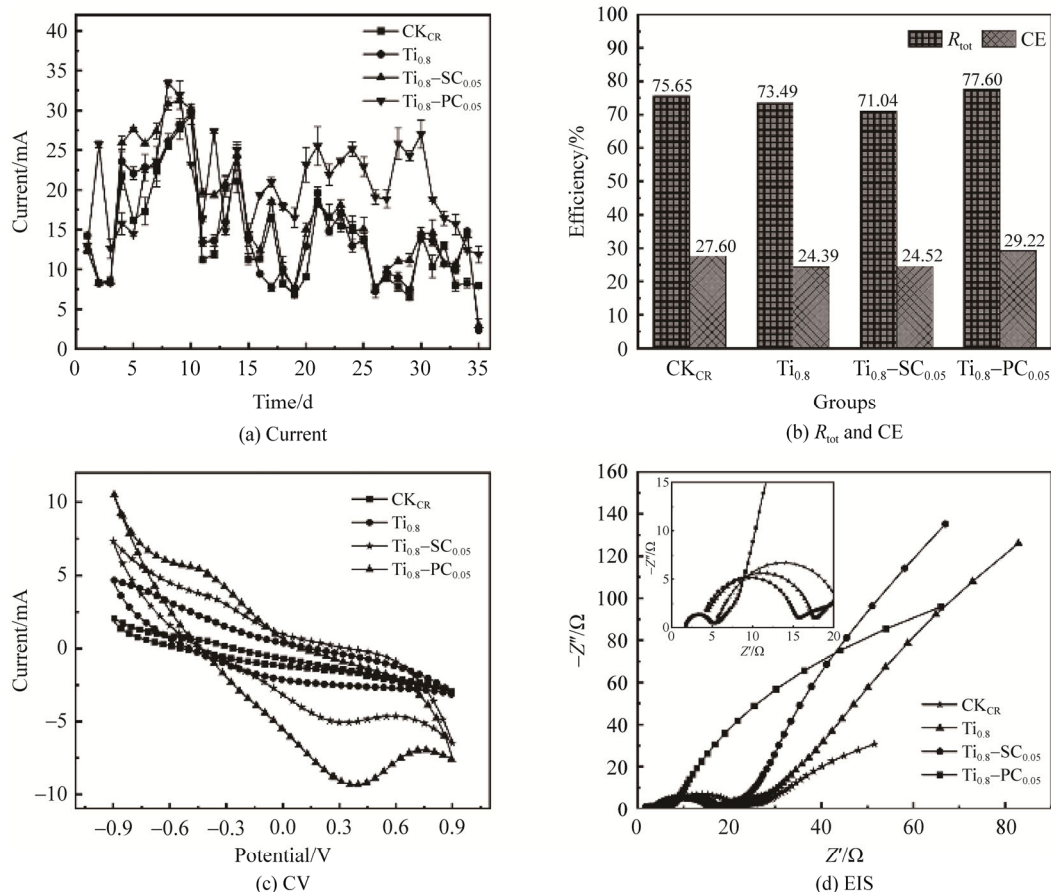


Fig. 5 Current generation, R_{tot} and CE, CV curves, and EIS curves of MEC-AD system with AVW-SC and AVW-PC as electrode

Table 2 Fit the values of the EIS Nyquist diagram with the relevant elements

Groups	R_{ct}/Ω	Error/%	R_s/Ω	Error/%
CK_{CR}	16.80	0.88	5.43	1.20
$\text{Ti}_{0.8}$	12.64	0.82	4.15	1.45
$\text{Ti}_{0.8}\text{-SC}_{0.05}$	12.01	0.85	3.63	1.91
$\text{Ti}_{0.8}\text{-PC}_{0.05}$	3.00	0.96	1.70	0.68

3.3 The effect of AVW-PC accelerants and electrodes on MEC-AD system

Two kinds of aloe vera waste-derived carbon, AVW-SC and AVW-PC were coated on the Ti mesh, which were used as electrode in MEC-AD system. All experimental groups in Fig. 6(a) showed similar gas production trends throughout the 35 d digestion phase.

The first peak appeared in 8–11 d, and the second peak appeared in 19–23 d. Compared with CK_{CR} and $\text{Ti}_{0.8}$, $\text{Ti}_{0.8}\text{-SC}_{0.05}$ has a higher gas production peak. To determine the optimal loading quantity of AVW-PC on the MEC electrode, different amounts of AVW-PC were coated Ti mesh. As an electrode of the MEC-AD system, its effects on the AD performance were examined. Two daily biogas production peaks appear on the 9–12 d and the 19–25 d in Fig. 6(b), respectively. Based on the above experiments, different concentrations of AVW-PC, as an accelerant, were added into MEC-AD system with the $\text{Ti}_{0.8}\text{-PC}_{0.1}$ electrode. The biogas production peak of each group with AVW-PC accelerant is obviously earlier than that of the $\text{Ti}_{0.8}\text{-PC}_{0.1}$ group without AVW-PC accelerant in Fig. 6(c), indicating that when AVW-PC is used as both an electrode and an accelerant, the digestion cycle can be shortened.

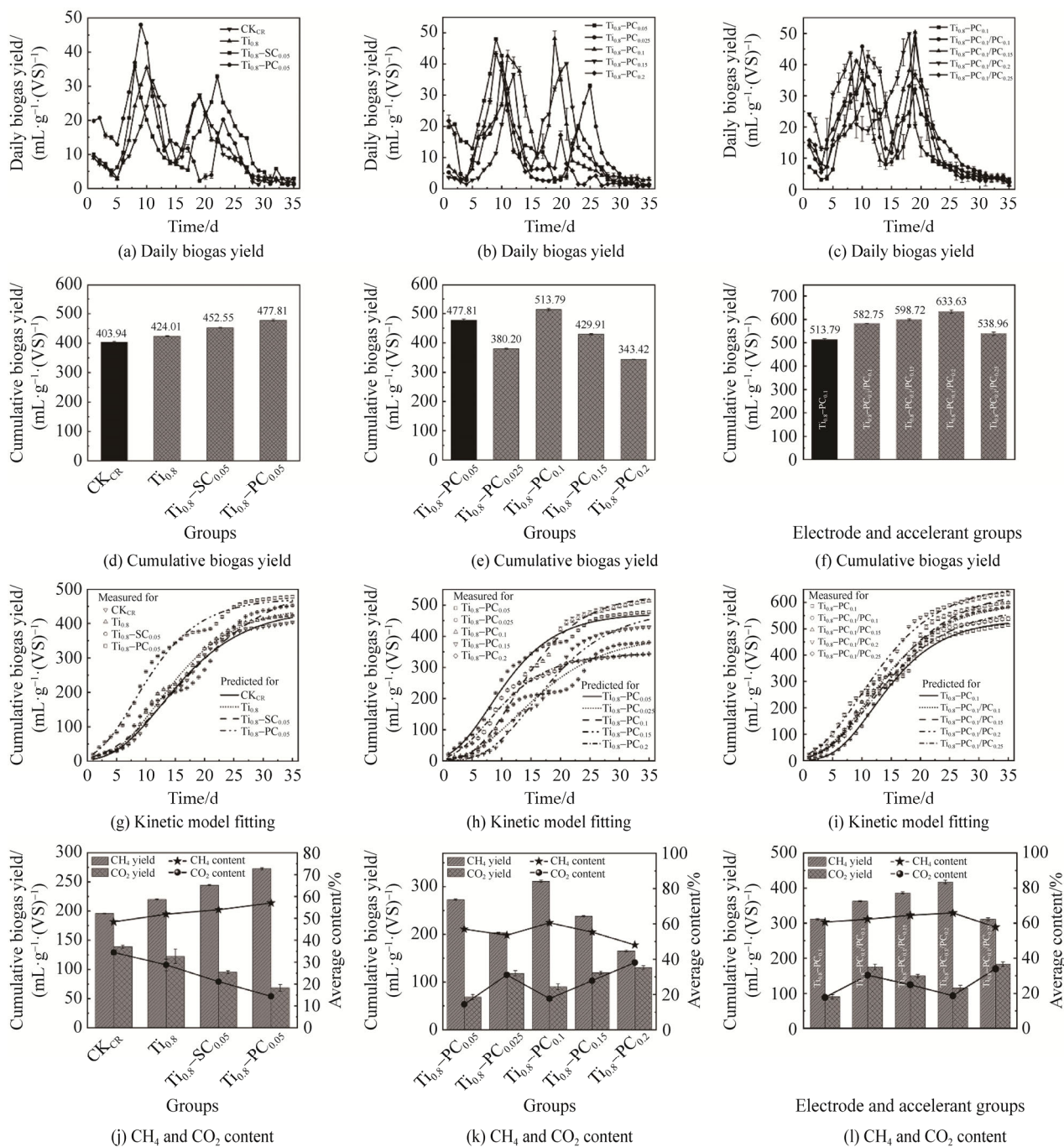


Fig. 6 Daily output of biogas, accumulated biogas yield, kinetic model fitting for cumulative biogas production, the yield and content of CH₄ and CO₂ of different electrode and accelerator groups

In addition, Ti_{0.8}-PC_{0.05} and Ti_{0.8}-SC_{0.05} groups, with Ti mesh coated with AVW-PC and AVW-SC as the electrode of MEC-AD, obtained larger biogas yields (477.81, 452.55 mL/g VS) in Fig. 6(d), 18.29% and 12.03% higher than that of CK_{CR} group, respectively. The Ti_{0.8}-PC_{0.1} group has a higher cumulative biogas production of 513.79 mL/g VS in Fig. 6(e). In Fig. 6(f), the Ti_{0.8}-PC_{0.1}/PC_{0.2} group managed to obtain a value of 633.63 mL/g VS. This result indicates a significant achievement for this group. The volume of 633.63 mL/g VS showcases the effectiveness and potential of the

Ti_{0.8}-PC_{0.1}/PC_{0.2} group in the context of the experiment being presented.

In general, the Gompertz model fits well with the data obtained from the experiment, and the R^2 values are 0.9876–0.9954 in Fig. 6(g). The fitting outcomes indicated that the Ti_{0.8}-PC_{0.05} group had gas production potential (P_m , 473.63 mL/g VS) and gas production (R_m , 28.11 mL/g VS/d), followed by Ti_{0.8}-SC_{0.05} (531.53 mL/g VS, 18.90 mL/g VS/d), Ti_{0.8} (450.36 mL/g VS, 22.43 mL/g VS/d), and CK_{CR} (442.95 mL/g VS, 20.61 mL/g VS/d). In Fig. 6(h), correlation coefficient (R^2) of the electrode

experimental group under different coating amounts was in the range of 0.9853–0.9970, which verified the rationality of the measured biogas production.

Additionally, the correlation coefficient (R^2) of AVW-PC as electrode and accelerant was between 0.9946–0.9970 in Fig. 6(i) (Tables 3–6).

Table 3 Kinetic parameters statistics of different voltage groups

Group 1	Measured cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	P_m /(mL·g ⁻¹ ·(VS) ⁻¹)	R_m /(mL·d·g ⁻¹ ·(VS) ⁻¹)	λ	Predicted cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	R^2
Ti ₀	313.79±5.10	358.28±7.47	14.31±0.36	5.27±0.26	321.65±7.85	0.9958
Ti _{0.4}	337.63±4.06	402.28±11.84	15.32±0.44	6.43±0.31	349.24±11.61	0.9942
Ti _{0.6}	365.21±5.24	459.24±17.98	15.77±0.46	6.38±0.34	380.55±15.33	0.9927
Ti _{0.8}	424.01±1.37	450.36±9.86	22.43±0.59	4.45±0.18	431.23±7.22	0.9947
Ti _{1.2}	387.80±3.43	448.33±0.78	22.44±0.70	4.33±0.28	429.94±5.93	0.9946

Table 4 Kinetic parameters statistics of different electrode groups

Group 2	Measured cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	P_m /(mL·g ⁻¹ ·(VS) ⁻¹)	R_m /(mL·d·g ⁻¹ ·(VS) ⁻¹)	λ	Predicted cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	R^2
CK _{CR}	403.94±2.43	442.95±9.81	20.61±0.49	4.69±0.17	417.63±13.69	0.9954
Ti _{0.8}	424.01±1.37	450.36±9.86	22.43±0.59	4.45±0.18	431.23±7.22	0.9947
Ti _{0.8} -PC _{0.05}	477.81±3.55	473.63±7.24	28.11±0.85	1.78±0.18	467.60±10.21	0.9945
Ti _{0.8} -SC _{0.05}	452.55±1.31	531.53±26.60	18.90±0.58	4.05±0.31	463.76±11.21	0.9876

Table 5 Kinetic parameters statistics of different AVW-PC electrode groups

Group 3	Measured cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	P_m /(mL·g ⁻¹ ·(VS) ⁻¹)	R_m /(mL·d·g ⁻¹ ·(VS) ⁻¹)	λ	Predicted cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	R^2
Ti _{0.8} -PC _{0.025}	380.20±1.41	402.54±17.13	18.24±0.85	4.65±0.41	377.33±2.87	0.9853
Ti _{0.8} -PC _{0.05}	477.81±3.55	473.63±7.24	28.11±0.85	1.78±0.18	467.60±10.21	0.9945
Ti _{0.8} -PC _{0.1}	513.79±3.97	535.72±8.32	30.40±0.66	5.38±0.13	520.83±7.04	0.9970
Ti _{0.8} -PC _{0.15}	429.91±2.53	487.46±15.83	23.66±0.70	7.49±0.21	453.58±23.67	0.9924
Ti _{0.8} -PC _{0.2}	343.42±0.81	345.09±6.56	21.79±0.93	1.88±0.24	341.91±1.50	0.9896

Table 6 Kinetic parameters statistics of different electrode and accelerant groups

Group 4	Measured cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	P_m /(mL·g ⁻¹ ·(VS) ⁻¹)	R_m /(mL·d·g ⁻¹ ·(VS) ⁻¹)	λ	Predicted cumulative biogas yield/(mL·g ⁻¹ ·(VS) ⁻¹)	R^2
Ti _{0.8} -PC _{0.1}	513.79±3.97	535.72±8.32	30.40±0.66	5.38±0.13	520.83±7.04	0.9970
Ti _{0.8} -PC _{0.1} /PC _{0.1}	582.75±1.05	601.62±9.35	29.94±0.59	3.32±0.13	579.53±3.22	0.9970
Ti _{0.8} -PC _{0.1} /PC _{0.15}	598.72±4.19	648.00±14.87	29.82±0.75	3.63±0.18	614.10±15.38	0.9946
Ti _{0.8} -PC _{0.1} /PC _{0.2}	633.63±6.86	665.89±9.98	32.60±0.69	2.19±0.15	643.28±9.65	0.9966
Ti _{0.8} -PC _{0.1} /PC _{0.25}	538.96±7.15	578.64±9.30	27.71±0.55	3.51±0.14	553.14±14.18	0.9969

Fig. 6(j) shows the CH₄ content and cumulative CH₄ yield of Ti_{0.8}-SC_{0.05} (53.94%, 244.35 mL/g VS) and Ti_{0.8}-PC_{0.05} (57.04%, 272.64 mL/g VS), higher than Ti_{0.8} (51.90%, 220.08 mL/g VS) and CK_{CR} (48.40%, 195.68 mL/g VS). Clearly, both AVW-SC and AVW-PC as electrodes can enhance the biogas production performance for the MEC-AD system. Compared with AVW-SC, AVW-PC can further improve the biogas production and methane content, so it could be more suitable as the electrode material. By contrast, it can be found in Fig. 6(k) that the Ti_{0.8}-PC_{0.1} group achieved optimal CH₄ content (60.59%) and cumulative CH₄ production (311.22 mL/g VS), followed by the

Ti_{0.8}-PC_{0.15} (55.33%, 238.17 mL/g VS), Ti_{0.8}-PC_{0.025} (53.56%, 203.54 mL/g VS), and Ti_{0.8}-PC_{0.2} (48.08%, 165.49 mL/g VS) group. This demonstrates that the Ti_{0.8}-PC_{0.1} group is more helpful in improving the gas production effectiveness for the MEC-AD system. In Fig. 6(l), Ti_{0.8}-PC_{0.1}/PC_{0.2} group obtained the highest CH₄ content (65.85%) and cumulative CH₄ production (417.45 mL/g VS), higher than Ti_{0.8}-PC_{0.1} (60.59%, 311.22 mL/g VS), Ti_{0.8}-PC_{0.1}/PC_{0.1} (62.22%, 362.58 mL/g VS), Ti_{0.8}-PC_{0.1}/PC_{0.15} (64.50%, 386.18 mL/g VS), and Ti_{0.8}-PC_{0.1}/PC_{0.25} (57.76%, 311.28 mL/g VS). Meanwhile, during the entire digestion process, the highest methane production in

the $\text{Ti}_{0.8}\text{-PC}_{0.1}/\text{PC}_{0.2}$ groups corresponded to a lower CO_2 content of 18.65%. The lower CO_2 concentration may be correlated with the excellent CO_2 adsorption and capture capacity, electron conductivity, and surface area of AVW-PC, thus promoting the process of CO_2 conversion to CH_4 ^[32]. Therefore, the AVW-PC serving as

both an electrode and an accelerant could significantly increase the biogas yield of MEC-AD system, which may be due to AVW-PC improving redox environment and microbial metabolism^[7, 16]. In addition, the methane content and yield with different electrodes were collected as shown in Table 7.

Table 7 Comparison of methane yield from MEC-AD systems with different electrode materials or accelerants

AD substrate	Cathode	Anode	Accelerant	Temperature/°C	Applied voltage/V	CH_4 contents/%	CH_4 yield/ ($\text{m}^3 \text{CH}_4/\text{kg COD}$)	Reference
Food waste	Stainless steel	Graphite plates		35	0.9	71.90	NG	[35]
Pig slurry+	Granular	Carbon felt		23±2	0.8	60.00	NG	[36]
Granular sludge	graphite							
Activated sludge+	Carbon	Carbon fiber felt		35	0.9	63.53*	0.29*	[37]
pig manure	fiber felt							
Anaerobic sludge+	Carbon felt	Carbon felt		35	NG	48.52	NG	[38]
pig manure	composite	composite						
Dairy manure	Stainless-steel cylinder	Graphite plates		35	1.2	63.00	NG	[39]
Sludge+Sodium acetate	Nickel screen	Carbon fiber brush		35	0.8	93.60	0.26	[40]
Sludge	Carbon brush	Carbon brush		35	0.8	NG	0.21*	[41]
Concentrated sludge	Carbon cloth	Carbon brush	Culture medium	35	0.6	45.60	NG	[42]
Straw	Carbon cloth	Carbon brush		NG	1	87.00	NG	[43]
Maize straw	Ti-AC	Ti-AC		35	4	64.50	0.27*	[44]
Phosphate buffer solution	Ni/Co-NC	Carbon fiber brushes		37±1	0.6	NG	0.34	[10]
Cattle manure+	Carbon rod	Carbon rod	CBC	36±1	0.6	63.59	0.19*	[7]
Aloe vera								
Cattle manure+	Carbon rod	Carbon rod	BP and MBP	36±1	0.6	62.50	0.20*	[45]
Aloe vera								
Cattle manure+	Carbon rod	Carbon rod	MXene	36±1	0.6	69.80	0.30	[15]
Aloe vera								
Cattle manure+	$\text{Ti}_{0.8}\text{-PC}_{0.1}$	$\text{Ti}_{0.8}\text{-PC}_{0.1}$	AVW-PC	36±1	0.8	65.85	0.28	This work
Aloe vera waste								

*Calculated from the given data. AC: Activated carbon. NG: Not given. CBC: Coconut-shell-derived bio-based carbon. BP: Black phosphorus. MBP: BP modified by hydrogen peroxide. AVW-PC: Aloe vera waste-derived porous carbon.

Compared with reported electrode or accelerant, the AVW-PC can produce more methane, indicating that AVW-PC as electrode and accelerant has a good improvement effect on the methane production. According to the economic analysis in the previous literature^[33], by comparing the cost of these accelerants and electrodes to the income generated from the increased biogas production, these accelerants and electrodes are not cost-effective and counterbalanced. But AVW-PC accelerant and electrode can reduce TCOD and solids content, resulting in lower costs for subsequent digestion treatment. The digestate can act as one component of a compound fertilizer, offering an option to reduce the cost of accelerants. In large-scale biogas plants, utilizing the biogas generated for cogeneration can result in additional economic and environmental benefits. Furthermore, the government is introducing policies (*i.e.*, investment subsidies) to reduce the cost and tax burden

of large biogas plants, reflecting efforts to develop renewable energy and combat environmental pollution^[34]. In addition, industrializing AVW-PC also needs to take into account overcoming technical, economic and environmental barriers.

3.4 The influence of various concentrations of AVW-PC accelerants on MEC-AD system with AVW-PC as the electrode

In Fig. 7(a), the change trend of pH value is similar in each experimental group. In the whole digestion stage, the pH range was between 6.07–7.94. When the AVW-PC is used as the electrode for the MEC-AD ($\text{Ti}_{0.8}\text{-PC}_{0.05}$), the TCOD removal rates are 56.10% for $\text{Ti}_{0.8}\text{-PC}_{0.05}$ group, higher than $\text{Ti}_{0.8}\text{-SC}_{0.05}$ (53.71%), $\text{Ti}_{0.8}$ (47.62%), and CK_{CR} (40.45%) in Fig. 7(b). This indicated that the AVW-PC as the electrode is more suitable for organic material decomposition.

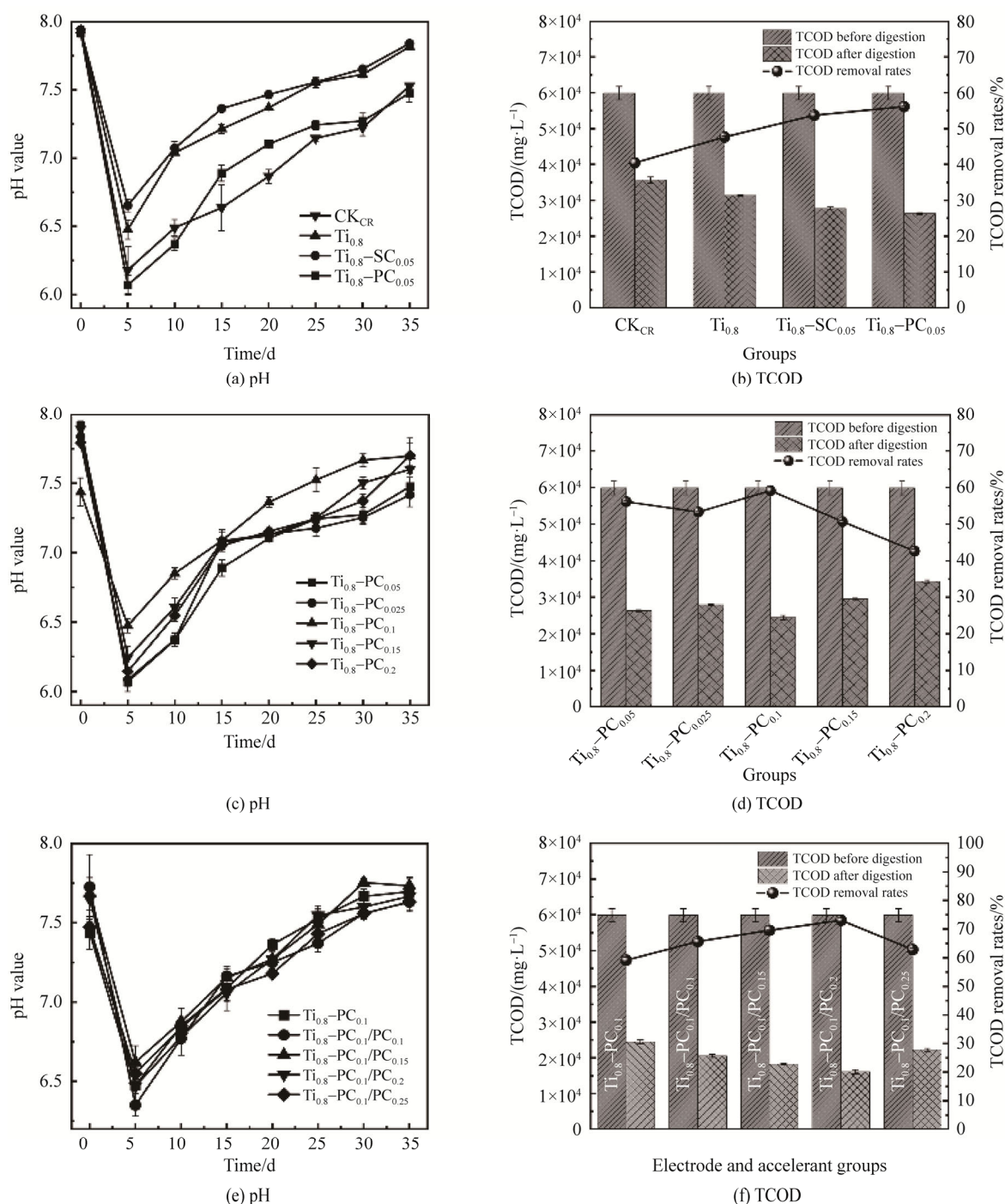


Fig. 7 pH, and TCOD of MEC-AD system with AVW-PC as electrode and accelerant

In addition, the pH variation was in the appropriate range during the whole digestion period in Fig. 7(c), and a higher TCOD removal rate of 59.15% was also obtained in Fig. 7(d) for Ti_{0.8}-PC_{0.1} group. Generally, the Ti_{0.8}-PC_{0.1} group exhibits optimal biogas production, substrate degradation, and methane content.

The pH value of each group was between 6.35 and 7.75 in Fig. 7(e), which is in line with the digestive environment. The TCOD degradation rate of before and after digestion in all reactors are shown in Fig. 7(f). Compared with Ti_{0.8}-PC_{0.1} (59.15%), higher TCOD

removal rate was obtained after adding accelerant. The Ti_{0.8}-PC_{0.1}/PC_{0.2} reached the highest TCOD degradation rate of 73.06%. This finding aligns with the cumulative biogas and CH₄ yield, suggesting that aloe vera waste-derived AVW-PC, serving both as an electrode and accelerant.

In Tables 8–11, the Ti mesh group in the MEC-AD system under different voltages obtained the TS removal rate of 28.45%–37.50% and the VS removal rate of 31.71%–48.55%. Of them, The Ti_{0.8} group is the best one in Table 8. The AVW-SC and AVW-PC coated Ti mesh

groups achieved the removal rate of TS (40.94%–44.23%) and VS (51.52%–53.17%) in the MEC–AD systems in Table 9, which surpass the $Ti_{0.8}$ (37.50%, 48.55%) and CK_{CR} (34.45%, 41.21%) groups. Notably, the $Ti_{0.8}$ – $PC_{0.05}$ group exhibits the removal of the TS (44.23%) and the VS (53.17%) in Table 9. Change in AVW–PC amount coated on the Ti mesh resulted in a substantial fluctuant TS and VS removal rate with a range of 36.58%–46.94% and 46.60%–55.31% in Table 10. Within this group, the $Ti_{0.8}$ – $PC_{0.1}$ achieves the highest removal rate, reaching 46.94% for TS and 55.31% for

VS. Further, employing the optimal AVW–PC electrode ($Ti_{0.8}$ – $PC_{0.1}$), various concentrations of AVW–PC as an accelerant in the MEC–AD systems yield notable TS removal rates of 46.94%–55.87% and VS removal of 55.31%–64.43% in Table 11. The maximum removal of TS and VS (55.87%, 64.43%) were seized in $Ti_{0.8}$ – $PC_{0.1}/PC_{0.2}$ group. The utilization of AVW–PC as the electrodes and accelerants notably enhanced the TS and VS removal efficiency, signifying an improved degradation of the organic substrate, which is consistent with the TCOD.

Table 8 TS and VS variations at the beginning and end of different voltage groups

Group 1	TS/(g·kg ⁻¹)		TS removal rates/%	VS/(g·kg ⁻¹)		VS removal rates/%
	Initial	Final		Initial	Final	
Ti_0	45.58±0.65	32.61±0.59	28.45±0.32	29.37±0.07	20.06±0.13	31.71±0.28
$Ti_{0.4}$	45.58±0.65	31.54±0.51	30.81±0.25	29.37±0.07	18.93±0.10	35.53±0.35
$Ti_{0.6}$	45.58±0.65	30.31±0.41	33.51±0.19	29.37±0.07	18.09±0.04	38.41±0.18
$Ti_{0.8}$	45.58±0.65	28.49±0.53	37.50±0.27	29.37±0.07	15.11±0.16	48.55±0.41
$Ti_{1.2}$	45.58±0.65	29.41±0.63	35.48±0.48	29.37±0.07	17.37±0.03	40.84±0.20

Table 9 TS and VS variations at the beginning and end of different electrode groups

Group 2	TS/(g·kg ⁻¹)		TS removal rates/%	VS/(g·kg ⁻¹)		VS removal rates/%
	Initial	Final		Initial	Final	
CK_{CR}	45.58±0.65	29.88±0.48	34.45±0.16	29.37±0.07	17.26±0.04	41.21±0.26
$Ti_{0.8}$	45.58±0.65	28.49±0.53	37.50±0.27	29.37±0.07	15.11±0.16	48.55±0.41
$Ti_{0.8}$ – $SC_{0.05}$	45.58±0.65	26.92±0.80	40.94±0.95	29.37±0.07	14.24±0.02	51.52±0.13
$Ti_{0.8}$ – $PC_{0.05}$	45.58±0.65	25.42±0.47	44.23±0.27	29.37±0.07	13.75±0.15	53.17±0.43

Table 10 TS and VS variations at the beginning and end of different AVW–PC electrode groups

Group 3	TS/(g·kg ⁻¹)		TS removal rates/%	VS/(g·kg ⁻¹)		VS removal rates/%
	Initial	Final		Initial	Final	
$Ti_{0.8}$ – $PC_{0.025}$	45.58±0.65	26.70±0.26	41.42±0.28	29.37±0.07	14.40±0.13	50.98±0.55
$Ti_{0.8}$ – $PC_{0.05}$	45.58±0.65	25.42±0.47	44.23±0.27	29.37±0.07	13.75±0.15	53.17±0.43
$Ti_{0.8}$ – $PC_{0.1}$	45.58±0.65	24.18±0.34	46.94±0.62	29.37±0.07	13.12±0.06	55.31±0.18
$Ti_{0.8}$ – $PC_{0.15}$	45.58±0.65	28.02±0.33	38.52±0.19	29.37±0.07	15.12±0.07	48.50±0.17
$Ti_{0.8}$ – $PC_{0.2}$	45.58±0.65	28.91±0.45	36.58±0.13	29.37±0.07	15.68±0.06	46.60±0.31

Table 11 TS and VS variations at the beginning and end of different electrode and accelerant groups

Group 4	TS/(g·kg ⁻¹)		TS removal rates/%	VS/(g·kg ⁻¹)		VS removal rates/%
	Initial	Final		Initial	Final	
$Ti_{0.8}$ – $PC_{0.1}$	45.58±0.65	24.18±0.34	46.94±0.62	29.37±0.07	13.12±0.06	55.31±0.18
$Ti_{0.8}$ – $PC_{0.1}/PC_{0.1}$	45.58±0.65	25.00±4.47	51.18±1.01	29.37±0.07	11.54±0.14	60.72±0.39
$Ti_{0.8}$ – $PC_{0.1}/PC_{0.15}$	45.58±0.65	24.18±5.11	53.08±0.28	29.37±0.07	11.06±0.09	62.33±0.22
$Ti_{0.8}$ – $PC_{0.1}/PC_{0.2}$	45.58±0.65	23.20±5.42	55.87±0.61	29.37±0.07	10.44±0.32	64.43±1.16
$Ti_{0.8}$ – $PC_{0.1}/PC_{0.25}$	45.58±0.65	26.00±4.31	48.25±0.16	29.37±0.07	12.38±0.22	57.83±0.68

3.5 Thermal characteristics and digestate fertilizer

3.5.1 Thermal stabilization characteristics of the digestates
Digestate stability was examined using thermal analysis methods such as thermogravimetry (TG), derivative thermogravimetry (DTG), and differential scanning calorimetry (DSC). The TG curve in Fig. 8(a) depicts mass loss during digestate pyrolysis as temperature increases. With rising temperature, mass loss will

increase attributable to the pyrolytic conversion of organic matter. The TG curve comprises three distinct phases. The first stage of the TG curve (below 200 °C) corresponds to near 75 °C in Fig. 8(b). The mass loss of the digestion residue is mainly due to the liberation of water, including the crystallization water and the absorbed water in the digestate. The stage from 200 °C to 400 °C corresponds to the peak seen on the DTG

(Fig. 8(b)), which is mainly caused by the combustion of organics including saccharide. The temperature above 400 °C is associated with the third summit of the DTG curve in the range of 430 to 450 °C and the fourth apex ranging from 570 to 650 °C (Fig. 8(b)). The observed in the third stage is intricate organics, including aromatic structures and lignocellulose^[7, 46]. CK_{CR} group had a lower mass residue in TG curve and a higher peak in DTG curve, which proved that the digestate from

CK_{CR} had more biodegradable organic matter. The cumulative mass loss of CK_{CR}, Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2} in Fig. 8(c) were 53.12%, 49.79%, 49.22%, and 46.14%, respectively, indicating that the AVW-PC electrodes enhanced the substrate degradation and adding the AVW-PC accelerants furthermore reduced the biodegradable components in the digestate, which corresponded to the outcomes observed.

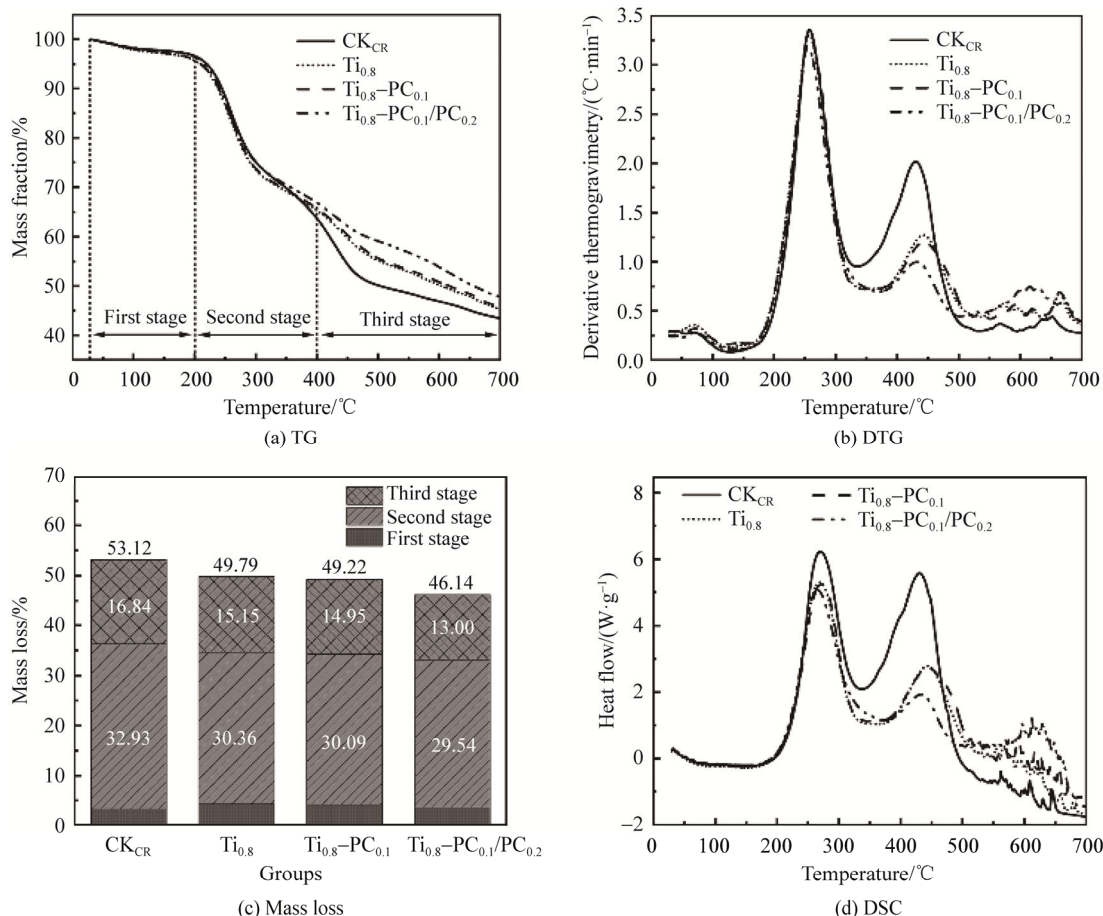


Fig. 8 TG, DTG, mass loss, and DSC graphs for the digestates of CK_{CR}, Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2} groups, which were selected from Groups 1–4 based on their optimal MEC-AD performance

The DSC curve depicted in Fig. 8(d) exhibits two peaks indicative of organic oxidation. At about 280 °C, the peak on the DSC curve corresponds to the degradable (including carbohydrates, cellulose and hemicellulose) components in the digestate, while the peak at about 430 °C is related to the refractory organic components (aromatic and lignin substances)^[45]. Compared with CK_{CR}, Ti_{0.8}, and Ti_{0.8}-PC_{0.1}, Ti_{0.8}-PC_{0.1}/PC_{0.2} showed lower exothermic peaks, indicating that these digestates were more stable. Consequently, the AVW-PC as electrodes and accelerants could enhance the decomposition of organic matter during MEC-AD process.

3.5.2 Fertility of the digestate The nutrient-rich residue can serve as a biofertilizer to enhance plant

growth and facilitate soil remediation, thereby enabling resource utilization. Table 12 presents the total nitrogen (TN), total phosphorus (TP), and total potassium (TK) contents in Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2}. The cumulative NPK nutrient content of organic fertilizer should be above 4% (40 g·kg⁻¹) in NY525–2021. The cumulative NPK in the digestates (38.87–42.30 g·kg⁻¹) in the present work far surpassed that obtained in the soil (3.10–4.50 g·kg⁻¹), and close to or higher than specified in the NY/T 525–2021 standard, which is a valuable ingredient in compound fertilizers for enhancing soil fertility. Besides, the content of TN (19.76, 18.06, and 19.13 g·kg⁻¹) in Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2} groups were higher than bio-organic fertilizer (17.10 g·kg⁻¹), while the content of TN (10.04, 13.14, and

12.94 g·kg⁻¹) and TP (9.06, 10.42, and 10.23 g·kg⁻¹) in Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2} groups were lower compared to that in the bio-organic fertilizer (18.00 and 25.40 g·kg⁻¹), suggesting potential use of this digestate as

nitrogen-rich fertilizer for nitrogen-deficient farmland. Also, the phosphorus-rich and potassium-rich accelerants should be taken into improving the application potential of the digestate.

Table 12 Content of TN, TK, and TP in digestate

Type	TN/(g·kg ⁻¹)	TK/(g·kg ⁻¹)	TP/(g·kg ⁻¹)	Cumulative NPK nutrient content/(g·kg ⁻¹)	Reference
Soil	0.15–0.20	0.10–0.15	0.06–0.10	3.10–4.50	[7]
Bio-organic fertilizer	17.10	18.00	25.40	60.50	[7]
NY/T 525–2021				40.00	Chinese Ministry of Agriculture
Ti _{0.8}	19.76±0.13	10.04±0.03	9.06±0.02	38.87±0.17	This work
Ti _{0.8} -PC _{0.1}	18.06±0.23	13.14±0.17	10.42±0.34	41.62±0.29	This work
Ti _{0.8} -PC _{0.1} /PC _{0.2}	19.13±0.53	12.94±0.04	10.23±0.33	42.30±0.82	This work

3.6 Examination of microbial community

Fig. 9 illustrates the microbial community composition in CK_{CR}, Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2}. In Figs. 9(a)–9(b), the bacterial community of CK_{CR} was mainly composed of *Bacteroidales* (18.63%), *Oscillospirales* (19.89%) and *Synergistales* (13.54%). At 0.8 V, the dominant bacteria in Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2} were *Bacteroidales* (11.53%, 21.21%, and 22.58%), *Pseudomonadales* (18.24%, 13.80%, and 17.77%) and *Oscillospirales* (13.40%, 13.72%, and 8.56%). These predominant orders comprise the principal functional hydrolytic and fermentative bacterial species within MEC–AD systems, capable of metabolizing organic materials to generate VFAs, CO₂, and H₂^[47, 49]. In Fig. 9(b), *Bacteroidales* is 21.21% for Ti_{0.8}-PC_{0.1} and 22.58% for Ti_{0.8}-PC_{0.1}/PC_{0.2}, and the abundance of *Pseudomonadales* is 13.80% for Ti_{0.8}-PC_{0.1} and 17.77% for Ti_{0.8}-PC_{0.1}/PC_{0.2}, respectively, higher than CK_{CR} (18.63% and 6.11%) and Ti_{0.8} (11.53% and 18.24%), indicating that the AVW–PC electrode and accelerant have a targeted enrichment effect on these hydrolytic bacteria.

Notably, the relative abundance of *Bacteroidales* in Ti_{0.8}-PC_{0.1}/PC_{0.2} (22.58%) was significantly higher than that in CK_{CR} (18.63%), Ti_{0.8} (11.53%) and Ti_{0.8}-PC_{0.1} (21.21%). In addition, according to Wang et al., *Bacteroidales* may contribute to acidogenesis, thereby reducing residual chemical oxygen demand (COD) levels in the digestate^[47]. Therefore, AVW–PC enhances the enrichment of *Bacteroidales*, which may be associated with the increased COD removal rate of Ti_{0.8}-PC_{0.1}/PC_{0.2} in Section 3.4. In addition, *Pseudomonadales* are dominant in anaerobic/anoxic systems, and investigations have unveiled its linkage of organic acids with long hydrocarbon chains^[49–50], indicating Ti_{0.8}, Ti_{0.8}-PC_{0.1} and Ti_{0.8}-PC_{0.1}/PC_{0.2} may promote the conversion of organics. Through the above analysis, the addition of AVW–PC as the electrode and accelerant of the MEC–AD system can make *Bacteroidales* and *Pseudomonadales* better enriched, thereby facilitating the degradation of organic matter.

Figs. 9(c)–9(d) depict that the predominant methanogens were *Methanobacteriaceae* (26.98% and

26.39%), *Methanospirillaceae* (19.48% and 23.64%), *norank_Bathyarchaeia* (24.35% and 15.11%), and *Methanosarcinaceae* (13.16% and 17.57%) in CK_{CR} and Ti_{0.8}. However, in Ti_{0.8}-PC_{0.1} and Ti_{0.8}-PC_{0.1}/PC_{0.2}, *Methanobacteriaceae* (21.23% and 28.96%), *Methanospirillaceae* (30.76% and 26.41%), *Methanosarcinaceae* (12.70% and 5.82%), and *Methanosaetaceae* (11.65% and 15.19%) were notably present. *Methanobacteriaceae* has been recognized to metabolize methanogenesis using formic acid rather than acetic acid as a substrate. It is involved in the degradation of propionic acid and the production of methane through interspecies hydrogen transfer (IHT)^[51]. Moreover, it has been proven that *Methanobacteriaceae* is widely found on the cathode biofilm. Its rapid development on the cathode biofilm would consume a large amount of hydrogen via the reaction ($\text{①}4\text{H}_2+\text{CO}_2\rightarrow\text{CH}_4+2\text{H}_2\text{O}$)^[51–52], further indicating its participation in the IHT process. The Ti_{0.8}-PC_{0.1}/PC_{0.2} group had a higher relative abundance (28.96%) of *Methanobacteriaceae* than the CK_{CR} (26.98%), Ti_{0.8} (26.39%), and Ti_{0.8}-PC_{0.1} (21.23%), indicating that the AVW–PC as electrode and accelerant favored the enrichment of the *Methanobacteriaceae*.

In addition, *Methanospirillaceae* is another proven methanogen with IHT ability, which is the most active player involved in methanogenesis. It was the dominant archaea of the hydrogenotrophic methanogenesis pathway, which could reduce CO₂ to CH₄ by consuming H₂^[53–54]. *Methanospirillaceae* is 23.64% for Ti_{0.8}, 30.76% for Ti_{0.8}-PC_{0.1} and 26.41% for Ti_{0.8}-PC_{0.1}/PC_{0.2}, higher than that of CK_{CR} (19.48%). This means that the methanogens can more actively participate in the IHT process for Ti_{0.8}, Ti_{0.8}-PC_{0.1} and Ti_{0.8}-PC_{0.1}/PC_{0.2} groups than the CK_{CR}. Studies have shown that the same dominance of *Methanosarcinaceae* and *Methanobacteriales* during AD of manure^[55]. Methanogen dynamic was shown by dominance of *Methanosarcinaceae* for acetotrophic methanogens (IAT). *Methanosarcinaceae*, as a representative of acetotrophic methanogens, can utilize a range of organic substrates for methane production ($\text{②CH}_3\text{COO}^-+\text{H}^+\rightarrow\text{CH}_4+\text{CO}_2$) in the IAT process^[55]. Archaeal community structures at the family level

confirm that *Methanosarcinaceae* was enriched in CK_{CR} (13.16%), Ti_{0.8} (17.57%), Ti_{0.8}-PC_{0.1} (12.70%) and Ti_{0.8}-PC_{0.1}/PC_{0.2} (5.82%) groups, which will promote the acetic acid ion transformation and methane production. More studies have found that *Methanosaetaceae*, a potential bioaugmented methanogen, is a typical microorganism involved in DIET ($3\text{H}^+ + 8\text{e}^- + \text{CO}_2 \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}$)^[20, 51]. As some studies indicate, real-time PCR analysis confirms that the *Methanosaetaceae* family, a key methanogen because of its ability to

facilitate direct interspecies electron transfer when attached to biochar, preferentially attaches to biochar^[56]. Therefore, *Methanosaetaceae* can serve as the dominant bacterial that produces methane through the DIET pathway. It should be noted that the abundance of *Methanosaetaceae* in the Ti_{0.8}-PC_{0.1}/PC_{0.2} group (15.19%) is higher than that in the other three groups. This means that the AVW-PC is used as both electrode and accelerant (Ti_{0.8}-PC_{0.1}/PC_{0.2}) at 0.8 V, the methanogens involved in the DIET process can be enriched.

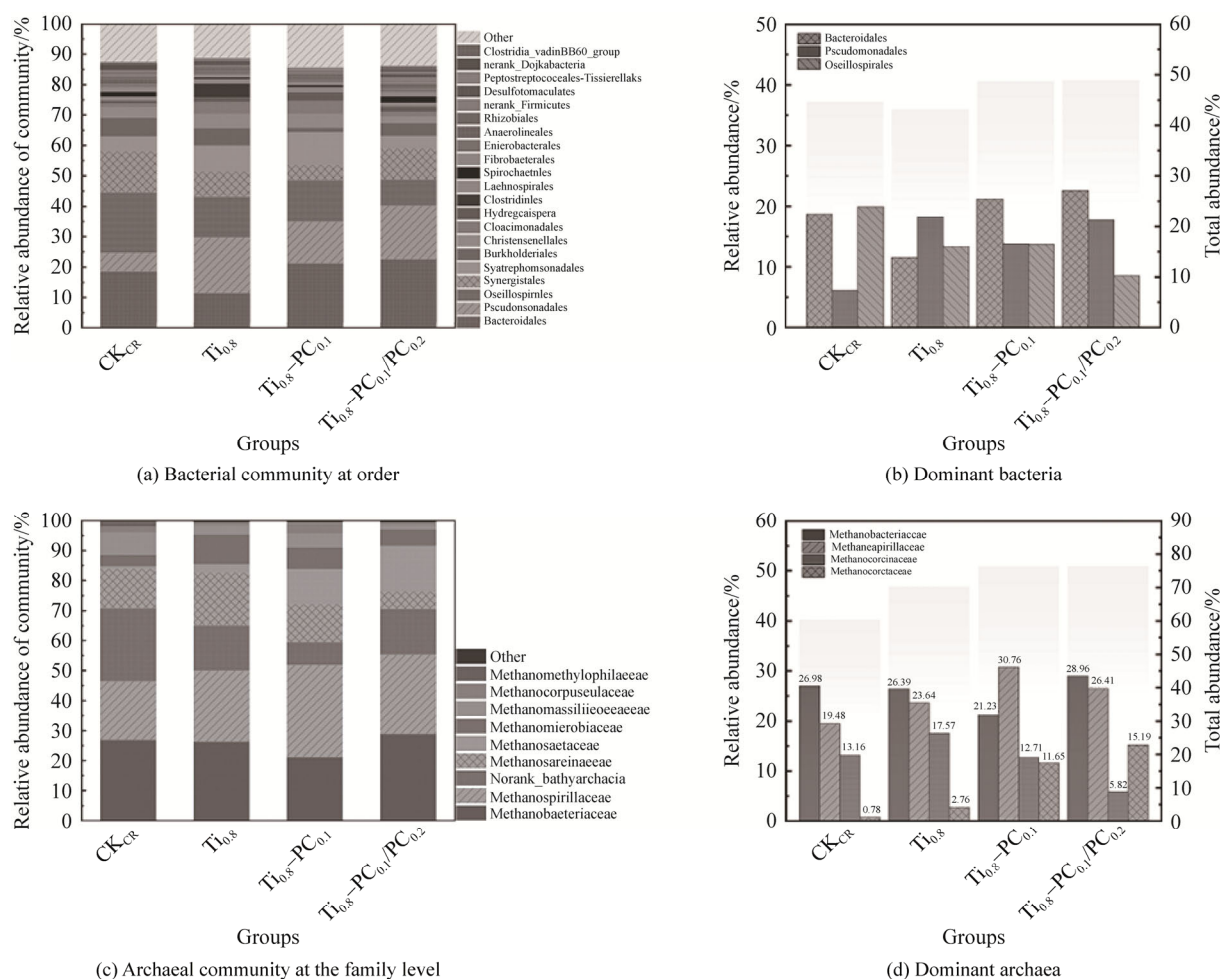


Fig. 9 Microbial community analysis of CK_{CR}, Ti_{0.8}, Ti_{0.8}-PC_{0.1}, and Ti_{0.8}-PC_{0.1}/PC_{0.2}

Overall, at a voltage of 0.8 V, the addition of AVW-PC as an electrode and accelerant, the abundances of *Bacteroidales*, *Pseudomonadales*, *Oscillospirales* and the abundances of *Methanobacteriaceae*, *Methanospirillaceae*, *Methanosarcinaceae*, *Methanosaetaceae*, participated in the IHT, IAT, and DIET process, were obviously riched, confirming the important role of the AVW-PC and the rich microorganisms.

3.7 Comprehending intensified methanogenesis in MEC-AD with AVW-PC as electrode and accelerant

1) AVW-PC as an electrode of the MEC-AD system Fig. 10 illustrates how to increase methane

production using AVW-PC as the electrode and acceleration. Electrochemically active bacteria are crucial for the extracellular electron transfer process, which is highly significant in microbial electrochemistry^[57]. As depicted in Fig. 10(a), these bacteria collaborate to decompose organic materials on the positive pole, causing the generation of electric charges, hydrogen ions, CO₂. Electrons are conveyed to the negative pole through an outside circuit. Simultaneously, the digestates carry hydrogen ions and CO₂. The negative pole receives electrons via an external circuit, and the digestates deliver H⁺ and CO₂. Methanogens present at the cathode catalyze the conversion of CO₂ with electrons and H⁺ ions into CH₄. The hydrogenotrophic methanogens

directly convert carbon dioxide and hydrogen gas to methane gas, while microorganisms that produce hydrogen at the cathode produce electron-catalyzed hydrogen directly^[53]. Acetotrophic methanogens make use of acetic acid and H^+ to generate CH_4 , while acetate-generating bacteria use electrons from the cathode to convert CO_2 into acetic acid^[15]. Another method is DIET, which reduces CO_2 to CH_4 by

transferring electrons from electrodes or fermenting bacteria to methanogens^[15, 45].

When AVW-PC is used as an electrode, it's delineated in two ways, as illustrated in Fig. 10(a). The AVW-PC possesses a honeycomb three-dimensional interconnected structure (Fig. 2(b)), which not only enhanced adherence of microorganisms but also provides electron transfer channels.

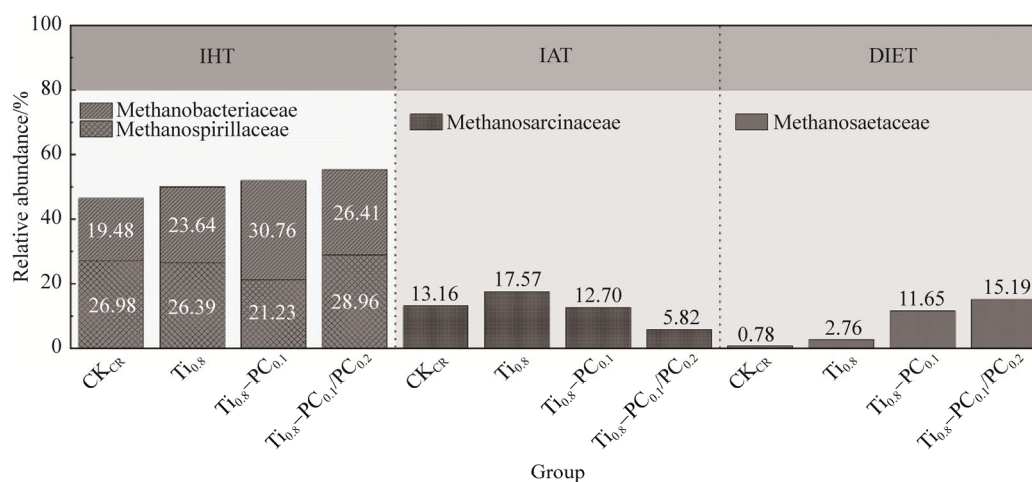
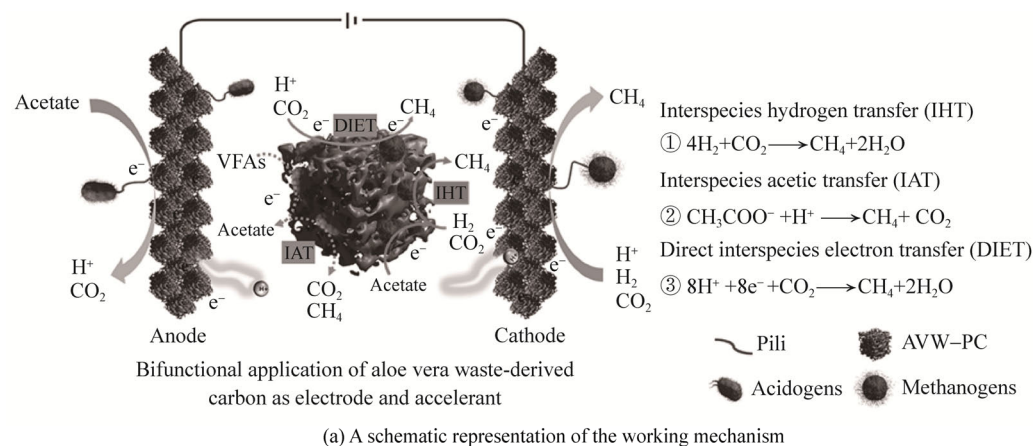


Fig. 10 A schematic representation of the working mechanism and alterations in syntrophic microorganisms mediating interspecific electron transfer in CK_{CR}, Ti_{0.8}, Ti_{0.8}-PC_{0.1} and Ti_{0.8}-PC_{0.1}/PC_{0.2} groups

During electron transfer, fermentative bacteria adhered to the anode or supplied externally serve as electron donors, while methanogens on the cathode function as electron acceptors. This facilitates effective electron transfer within the channels, thereby enhancing overall electron transfer efficiency. Concurrently, the AVW-PC electrode serves as a significant support medium for the attachment of organic matter. This promotes the decay of organic compound, leading to the electron generation process, H^+ , and CO_2 , subsequently fostering methane production through the IHT and IAT processes. The conductivity of the AVW-PC-coated Ti mesh electrode is also a crucial factor in promoting electron transfer. The highly conductive AVW-PC enhances the DIET process, thereby accelerating CH_4

production.

The experimental evidence in this study confirms and supports enhanced IHT and DIET processes. AVW-PC exhibits a good EEC ($0.75 \mu\text{mol} \cdot \text{e}^-/\text{g}$), indicating its potential as an electron transport carrier. Compared to CK_{CR} and pure Ti electrodes, the AVW-PC electrode (Ti_{0.8}-PC_{0.1} group) shows larger redox peaks in cyclic voltammetry (5.43 and -9.33 mA), confirming its improved redox activity. The larger capacitance in CV and smaller electrode internal resistance in EIS (3.00Ω) suggest that the AVW-PC electrode enhances electron transfer. Additionally, incorporating the AVW-PC electrode into the MEC-AD system enhances microbial enrichment involved in the IHT and DIET processes (Section 3.6). Relative abundances of *Bacteroidales*,

Pseudomonadales, and *Oscillospirales* increase with the AVW-PC electrode compared to CK_{CR} and pure Ti electrodes, promoting hydrolysis in the MEC-AD system. Moreover, increased relative abundances of *Methanobacteriaceae*, *Methanospirillaceae*, and *Methanosaetaceae* primarily enhance the IHT and DIET processes in the MEC-AD system. However, *Methanosarcinaceae*, associated with IAT, exhibits a relative abundance of 12.70% in the Ti_{0.8}-PC_{0.1} group. The increase in bacteria related to IHT (*Methanobacteriaceae* and *Methanospirillaceae*) and DIET (*Methanosaetaceae*) enhances IHT and DIET processes, potentially weakening the IAT process.

2) AVW-PC as the accelerant

The integration of AVW-PC as an accelerant led to an improvement in IHT, IAT, and DIET processes, elucidated from the perspective of microbial enrichment.

Initially, in the IAT process, *Methanosarcinaceae* enrichment significantly decreased from 12.70% in the Ti_{0.8}-PC_{0.1} group to 5.82% in the Ti_{0.8}-PC_{0.1}/PC_{0.2} group. This decline suggests reduced reliance on the IAT process with the addition of AVW-PC, as depicted in Fig. 10(b) (confirmed in Fig. 9), potentially linked to improved IHT and DIET processes. Subsequently, in the IHT process, *Methanobacteriaceae* and *Methanospirillaceae* enrichment in the Ti_{0.8}-PC_{0.1}/PC_{0.2} group surpassed that of the Ti_{0.8}-PC_{0.1} group, indicating further enhancement of the IHT process with the AVW-PC accelerant, as demonstrated in Fig. 10(b) (confirmed in Fig. 9). Thirdly, in the DIET process, *Methanosaetaceae* enrichment increased from 11.65% in the Ti_{0.8}-PC_{0.1} group to 15.19% in the Ti_{0.8}-PC_{0.1}/PC_{0.2} group (Fig. 10(b) (as confirmed in Fig. 9)), underscoring the pivotal role of AVW-PC in facilitating the DIET process. Based on experimental evidence^[2, 4, 7], AVW-PC has excellent electron exchange capabilities, improved the electrochemical performance of the MEC-AD system, AVW-PC and enriched methane-producing microorganisms (*Methanosaetaceae*) involved in the DIET process, indicating that AVW-PC can promote methane generation through DIET (Fig. 10). Overall, the utilization of AVW-PC as an accelerant strengthened the processes and pathways of methane production in the IHT and DIET processes, thereby fostering methane production.

The AVW-PC serves dual roles as both an electrode and an accelerant within the MEC-AD system, playing a pivotal role in its functionality. Functioning as an electrode, the AVW-PC adheres to microorganisms, thereby enhancing microbial activity and facilitating electron transfer in the IHT, IAT, and DIET processes. Furthermore, AVW-PC adsorbs the surrounding metabolites and mediates electron exchange between acidogens and methanogens, thereby fostering microbial enrichment and promoting substrate degradation. Consequently, the utilization of AVW-PC enhanced microbial attachment and microorganism growth, augmented the degradation of organic substrates,

accelerated electron migration. Eventually, the effectiveness and yield of methane synthesis were raised in the IAT, IHT, and DIET.

4 Conclusions

This work proves the successful carbonization of AVW-SC, and AVW-PC derived from aloe vera waste. The incorporation of aloe vera waste-derived carbon in the MEC-AD system, serving as both an electrode and an accelerant, shows its versatile functionality. The Ti_{0.8}-PC_{0.1}/PC_{0.2} group reached the maximum CH₄ production (417.45 mL/g VS) and TCOD removal rate (73.06%). The digestate of the Ti_{0.8}-PC_{0.1}/PC_{0.2} group exhibited good thermal stability (mass loss of 46.14%) and fertilizer efficiency (42.30 g·kg⁻¹), rendering it suitable for use as a fertilizer component. By serving as both an electrode and an accelerant, AVW-PC enhances hydrolysis efficiency by enriching *Bacteroidales*, *Pseudomonadales*, and *Oscillospirales*, while also promoting the IHT, IAT, and DIET processes through increased abundance of *Methanobacteriaceae*, *Methanospirillaceae*, *Methanosarcinaceae*, and *Methanosaetaceae* colonies. This study presents a promising strategy to enhance methane production using biomass-derived carbon (AVW-SC and AVW-PC) as electrodes and accelerants, contributing to the comprehensive and efficient utilization of aloe vera waste.

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MEC-AD Leveraging Derived Carbon for Energy-Efficient Methane Production: Insights into Electrodes, Accelerants, and Methanogenic Approaches

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Extended Abstract

Introduction The generation of biological wastes such as cow dung and aloe vera waste (AVW) causes a serious ecological pollution. The microbial electrolytic cell coupled with anaerobic digestion (MEC-AD) system can make a rational utilization of these biodegradable organic wastes, which is of vital importance for alleviating environmental deterioration and reducing resource waste. Electrode materials and accelerants are the two major factors that affect methane production in the MEC-AD system. They affect microbial attachment and electron transfer in the MEC-AD system. Bio-based carbon materials are carbon materials prepared from biomass as raw materials. They have characteristics such as a rich pore structure, good chemical stability, biocompatibility, and controllable surface properties, which can be used as accelerants and electrodes in the MEC-AD system to optimize its performance. This study was to investigate the influence of biomass-derived carbon as an electrode and accelerant on the performance of the MEC-AD system, and the mechanism for increasing the production of biogas and methane was also analyzed, thus providing a basis for the multifunctional application of biomass-derived carbon in the MEC-AD system.

Methods A series of experimental methods were adopted to study the MEC-AD system. Two types of bio-based carbon, *i.e.*, aloe vera waste derived spherical carbon (AVW-SC) and porous carbon (AVW-PC), were synthesized *via* hydrothermal carbonization.

The raw AVW material was washed with water, dried, ground, and subjected to hydrothermal treatment to obtain AVW–SC. After activating AVW–SC with KOH, it was carbonized in a tube furnace to obtain AVW–PC.

In the preparation of the electrodes, bio-based carbon (AVW–SC and AVW–PC) was mixed with 5% polytetrafluoroethylene powder in ethanol and deionized water, and then ground in a ball mill for 4 h to form a slurry. The slurry was evenly sprayed on the Ti mesh, dried and sintered in N₂ atmosphere at 360 °C to obtain Ti–SC and Ti–PC electrodes.

Four groups of experiments were conducted to determine the optimal voltage, compare different carbon electrodes, and explore the optimal coating amount. The MEC–AD reactor adopted 500 mL wide-mouthed glass bottles with a working volume of 400 mL. Each MEC–AD system received a co-substrate mixture of cattle dung and aloe vera waste and inoculum of sewage sludge in a mass ratio of 3:7. Afterward, they were placed at (36±1) °C for 35 d. The biogas was collected by a water displacement method.

The materials were analyzed by characterization techniques such as X-ray diffraction (XRD) and scanning electron microscopy (SEM), and electrochemical tests were conducted on different electrodes. The composition, pH, TS, VS, TCOD and nutrient content of biogas were analyzed by standard chemical methods. Microbial community analysis was conducted using high-throughput sequencing technology. The modified Gompertz model was adopted to predict the kinetic parameters, and the coulombic efficiency and methane recovery rate were calculated according to a specific formula.

Results and discussion The result shows that AVW–SC is spherical and closely aggregated, while AVW–PC has a three-dimensional network structure, with average pore diameter of 9.77 nm. The electron exchange capacity (EEC) of AVW–PC (*i.e.*, 0.75 μmol·e⁻/g) is higher than that of AVW–SC (*i.e.*, 0.15 μmol·e⁻/g), indicating a better electron exchange capacity. These results indicate that AVW–PC provides more substrate and bacteria accumulation sites, and has better electron-donating and electron-accepting ability, thus improving the digestion efficiency.

In the MEC–AD system, using Ti mesh as an electrode, the effect of different voltages (*i.e.*, 0, 0.4, 0.6, 0.8 V and 1.2 V) on the system performance is investigated, obtaining the optimum biogas production and organic matter degradation rate at 0.8 V. AVW–SC and AVW–PC are respectively coated on Ti mesh as electrodes. The results show that the MEC–AD system with AVW–PC coated Ti mesh as the electrode has a better performance. The electrochemical analysis shows that the electrode coated with AVW–PC has a larger specific capacitance and a smaller charge transfer resistance, indicating that AVW–PC can improve the electrochemical properties and electron transfer ability of MEC–AD system. The influence of coating amount (*i.e.*, 0.025, 0.05, 0.10, 0.15, and 0.20 g) of AVW–PC on the MEC–AD system is investigated. At a coating amount of AVW–PC of 0.1 g, the cumulative biogas production and methane content of the Ti_{0.8}–PC_{0.1} group both reach the maximum values. Different doses of AVW–PC (*i.e.*, 0.10%, 0.15%, 0.20%, and 0.25%) are added as accelerants in Ti_{0.8}–PC_{0.1}. At the addition amount of AVW–PC of 0.20%, the Ti_{0.8}–PC_{0.1}/PC_{0.2} group performs the optimum biogas production (*i.e.*, 633.63 mL/g VS), methane content (*i.e.*, 65.85%), and total nutrient content of biogas residue (*i.e.*, 42.30 g/kg). In Ti_{0.8}–PC_{0.1}/PC_{0.2}, *Bacteroidales*, *Pseudomonadales*, *Oscillospirales*, *Methanobacteraceae*, *Methanospirillaceae*, *Methanosarcinaceae* and *Methanosaetaceae* significantly increase. The increase in microbial diversity promotes interspecific hydrogen transfer (IHT), interspecific acetic transfer (IAT), and direct interspecific electron transfer (DIET), thereby enhancing methanogenic efficiency.

Conclusions AVW–SC and AVW–PC were utilized as electrodes and accelerants to enhance methane yield in MEC–AD system. The Ti mesh electrode coated with different concentrations of AVW–PC achieved the optimal biogas production at 0.8 V. Specifically, the Ti_{0.8}–PC_{0.1} combination could generate the maximum total amount of biogas and methane proportion. The Ti_{0.8}–PC_{0.1}/PC_{0.2} combination exhibited the optimum performance (*i.e.*, biogas yield of 633.63 mL/g VS, methane content of 65.85% and total nutritional content of 42.30 g/kg). High abundances of *Bacteroidales*, *Pseudomonadales*, *Oscillospirales*, *Methanobacteraceae*, *Methanospirillaceae*, *Methanosarcinaceae*, and *Methanosaetaceae* appeared in the Ti_{0.8}–PC_{0.1}/PC_{0.2} group, compared to other groups. In addition, an increased microbial diversity led to an enhanced methane production through processes like DIET. This research could highlight the potential significance of AVW–PC as both electrode and accelerator for increasing methane production and provide a perspective for improving MEC–AD performance through multiple applications of biomass-derived carbon.

Keywords aloe vera waste; biomass-derived carbon; electrode; accelerant; biogas production