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Biochar addition supports high digestion performance and low membrane fouling rate in an anaerobic membrane bioreactor under low temperatures

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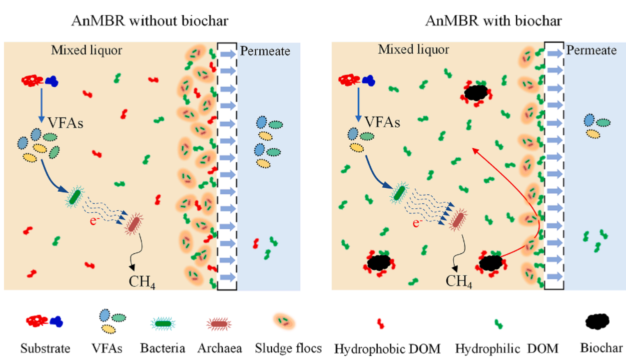
HIGHLIGHTS

- Biochar sustained high digestion performance and low membrane fouling of AnMBRs.
- Electron transfer system activity and VFAs degradation were enhanced by biochar.
- Biochar improved the sludge filtration property and decreased the hydrophobic DOM.
- Biochar decreased the cake/gel foulants ratio by mitigating cake layer formation.

GRAPHICAL ABSTRACT



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ABSTRACT

The enhancement effects of biochar to an anaerobic membrane bioreactor (AnMBR) treating sewage at low temperatures was investigated in this study through analyzing organics removal, digestion performance, mixed liquor properties, membrane resistance, and foulant compositions. The chemical oxygen demand (COD) removal efficiency and the COD converted to methane rate increased by more than 12.5% at 10 °C, mainly because of the promotion of biochar to volatile fatty acids degradation. Although biochar caused higher dissolved organic matter (DOM) concentration in the AnMBR, it improved the filtration property of the bulk sludge and absorbed the hydrophobic DOM. The decreased filtration resistance assisted by biochar leads to a prolonged membrane operation duration over 200%. Surface foulants, especially cake foulants, were largely mitigated by the enhanced scouring intensity of mixed liquor at the membrane surface, and hence, decreasing the cake/gel foulants ratio.

1. Introduction

Treating sewage using anaerobic membrane bioreactors (AnMBRs) to remove organics and recover chemical energy has attracted much attention recently (Ji et al., 2021; Nie et al., 2017). This process achieved high-rate oxygen demand (COD) removal efficiency (81–95%) and

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methane recovery efficiency (60–80%) in some pilot-scale cases (Kong et al., 2020a; Shin et al., 2014), with a low sludge yield of 0.05–0.22 g of volatile suspended solids per gram of COD removed (Kong et al., 2020b; Robles et al., 2020; Shin et al., 2014), showing merits in organics removal, energy recovery, and sludge disposal than aerobic and conventional anaerobic treatment processes (Chen et al., 2017b). However, obstacles and challenges still exist and require intensive study to facilitate widespread application. A short hydraulic retention time (HRT) needs to be sustained for high-rate microbial proliferation and methane recovery due to the low-strength of sewage (about 500 mg COD /L) (Watanabe et al., 2017). Under this condition, the low temperature of the sewage (5–28 °C) will restrain the digestion and membrane filtration performances, especially AnMBRs located in temperate or frigid regions (Lettinga, 2001; Vinardell et al., 2020).

Ho and Sung (2010) found that the COD removal efficiency decreased by 10% when the temperature decreased from 25 °C to 15 °C due to the suppressed methanogenic activity, similar results were also reported in other studies (Gao et al., 2014; Giménez et al., 2014). Under these conditions, membrane fouling was exacerbated due to the compensation of membrane interception to the decreased biological removal rate and the increased mixed liquor viscosity (Dev et al., 2019). Moreover, low temperatures also inhibit the hydrolysis of particulate COD, leading to a higher sludge yield (Giménez et al., 2014). Although these challenges have been thoroughly investigated and well understood, few practical protocols have been proposed to solve this unfavorable situation. Adjusting operational strategies, such as prolonging solids retention time and decreasing membrane flux, have been widely adopted, while the effectiveness of these measures is limited (Smith et al., 2013), and will lead to other issues (e.g., aggerated membrane fouling by high solids concentration, decreased volume loading, and increased investment). Using various additives, including adsorbents (Lei et al., 2019; Zhang et al., 2017), flocculants (Dong et al., 2015), non-adsorbing polymeric particles (Chen et al., 2017a), and some nanomaterials (Zhang et al., 2020), to optimize AnMBR operation have been comprehensively investigated. Among these additives, flocculants, and nanomaterials easily pass out accompanying sludge discharge and, therefore, need to be replenished regularly (Skouteris et al., 2015). In addition, these powder additives will also cause an increased sludge yield and potential ecological risks when being discharged into the environment.

Carbon-based materials produced from agricultural waste, including activated carbon and biochar, are environmentally friendly and can be easily recycled when produced in granulated form. The efficacy of activated carbon for enhancing anaerobic digestion and retarding membrane fouling has been investigated and demonstrated in previous studies (Hu and Stuckey, 2007; Yang et al., 2019). Compared with activated carbon, biochar can be easily produced at a lower pyrolysis temperature without activation, indicating that a much lower investment is required (Singh et al., 2017). In addition, biochar has a lower specific gravity than granular activated carbon, meaning that less energy is required for additive fluidization (Yargicoglu et al., 2015). Previous studies reported that biochar has comparable efficacy with commercial activated carbon in promoting anaerobic digestion (Li et al., 2018, 2020), and the feasibility of biochar in alleviating membrane fouling has been demonstrated in aerobic MBRs (Sima et al., 2017). However, the absorbability of biochar is much weaker than activated carbon because of its relatively low specific surface area (Wang et al., 2018a); therefore, the impact of biochar on the properties of the mixed liquor may differ from activated carbon. Although biochar produced by different kinds of biomass has been used in the anaerobic treatment processes, previous studies paid majority attention to the promotion of biochar to VFAs utilization and microbial evolution in high-strength waste treatment (Li et al., 2018; Sima et al., 2017; Wang et al., 2018b), their roles in low-strength sewage treatment at low temperatures, membrane fouling mitigation, and fouling layer compositions have not been comprehensively studied.

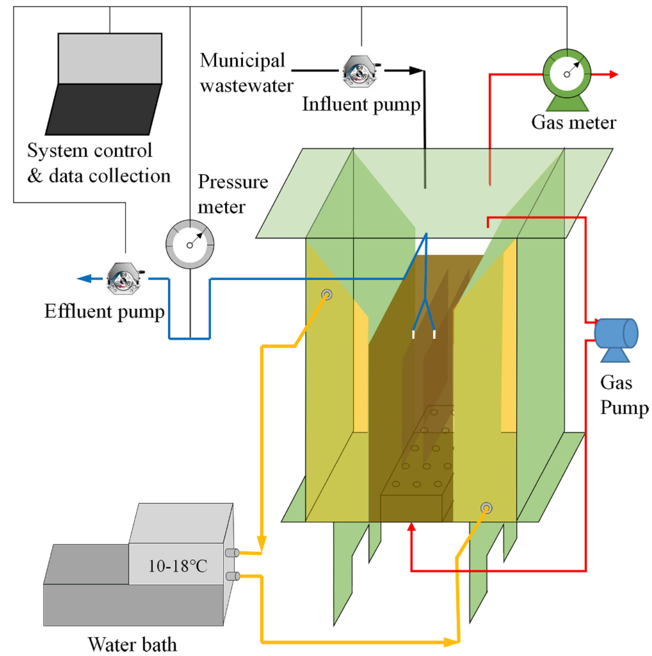

Fig. 1. The schematic diagram of the AnMBR system constructed in this study.

In this study, biochar prepared with low-cost corncobs through a simple production process was used to stimulate digestion performance and retard membrane fouling during anaerobic treatment of sewage using an AnMBR. The effects of biochar on digestion efficiency, COD removal, mixed liquor properties, and membrane fouling control were investigated and evaluated to examine the feasibility of mediating AnMBR operating at low temperatures by biochar.

2. Materials and methods

2.1. AnMBR set-up and operation

A submerged AnMBR reactor with an effective volume of 3.0 L (total volume of 5.0 L) was constructed and operated at 18 °C for over six months before this study. The reactor was fed with synthetic wastewater as in a previous study (Yang et al., 2019), and the total COD, total nitrogen, and total phosphorus were set at 500, 50, and 5 mg/L, respectively. Two polyvinylidene fluoride membrane modules with a pore size of 0.1 μm were set into the reactor. Feeding and drainage of the AnMBR were implemented using peristaltic pumps. The biogas generated from the anaerobic digestion in the AnMBR was collected and measured using a wet gas meter (LMF-1, Wale, China). Biogas in the headspace of the AnMBR was continuously recycled to the distribution box at the AnMBR bottom using a gas pump (VBY7506, Cheehie, China) with a circulation rate of 3.5 L/min to retard membrane fouling. Trans-membrane pressure (TMP) was monitored online using a pressure meter installed on the effluent line. The schematic diagram of the AnMBR can be seen in Fig. 1.

Two operating temperatures (18 °C and 10 °C) were adopted in this study for the AnMBR. At each operating temperature, the AnMBR was operated with (2 g/L-reactor) or without biochar separately, to investigate the impact of biochar on digestion and membrane fouling. When the membrane is fouled (TMP = 30 kPa) or the setpoint is reached, membranes in the AnMBR are replaced using new membranes. The detailed operating information is provided in Table 1.

2.2. Biochar preparation

Corncob granules were used as the biomass source for biochar preparation through pyrolysis. According to a previous study (Wang

Table 1
Operation performance of the AnMBR at different operating stages.

Operating stages	Operation conditions	Membrane aera (m^2) and average membrane flux (L/m^2/h)	Effluent COD (mg/L)	COD removal efficiency (%)	Acetate in effluent (mg/L)	Propionate in effluent (mg/L)	PN in effluent (mg/L)	PS in effluent (mg/L)
Phase I (Stage I)	18 °C, without biochar	0.07; 11.0	39.9 ± 16.9	90.5 ± 3.7	2.3 ± 0.8	<1.0	10.5 ± 2.0	1.3 ± 0.7
Phase II (Stage I)	18 °C, with biochar	0.07; 11.0	26.8 ± 3.2	94.4 ± 0.8	2.0 ± 0.9	<1.0	12.0 ± 2.6	0.9 ± 0.4
Phase III (Stage II)	18 → 10 °C, without biochar	0.07→0.12; 11.0 → 6.5	26.8 → 123.0	94.4 → 55.7	2.0 → 26.7	0.8 → 9.6	/	/
Phase IV (Stage III)	10 °C, without biochar	0.12; 6.5	122.0 ± 1.7	74.3 ± 7.6	39.8 ± 17.2	14.3 ± 4.7	15.7 ± 3.4	1.2 ± 0.2
Phase V (Stage III)	10 °C, with biochar	0.12; 6.5	62.9 ± 28.7	86.6 ± 5.2	13.6 ± 8.9	2.9 ± 3.2	14.1 ± 3.7	1.2 ± 0.6

"/" means data are not collected.

et al., 2020), a low pyrolysis temperature of 500 °C was taken in this study to reach the optimizing promotion of biochar to anaerobic digestion by generating abundant redox-active organic functional groups in biochar. The air-dried corncob was placed in a controlled atmosphere furnace (Therm, SAF, China) using pure nitrogen gas as the shielding gas. The heating rate was set as 16.6 °C /min; after 0.5 h of operation, the temperature reached 500 °C, and this temperature was maintained for 1.5 h. After the pyrolysis process, the biochar sample was cooled to room temperature and then sieved to uniform size fractions of 2.0–3.0 mm.

2.3. Samples collection

According to present knowledge, physical washing is capable of delaminating gel and cake foulants (Wang et al., 2008). So, the membrane samples were treated through water scouring to delaminate cake and gel foulants as the following procedure: The cake foulants were peeled off by scouring along the tangent plane using ultrapure water until the macroscopic sludge particles were removed, then the residual transparent gel layer foulants on the membrane were wiped off with a piece of sponge. Cake and gel foulants were collected separately after water scouring and sponge scrubbing, respectively, and pure water was added up to a certain volume (250 mL) to get the suspended liquid of gel and cake foulants. These suspended liquid samples were mixed well and used to analyze the components and amount of layer foulants. A portion of the suspended liquid was filtered using a syringe filter with a pore size of 0.45 μm, and the filtrate was collected as the liquid foulants of cake and gel layers.

Influent and effluent samples were collected every two days for direct COD analysis. The mixed liquor was centrifuged at 4500 rpm for 10 min, and the supernatant was filtered through a 0.45 μm membrane and collected as dissolved organic matter (DOM). The virgin and used biochar was cryoground and air dried at a low temperature (40 °C) for spectrographic and morphologic analysis.

2.4. DOM characterization through size-exclusion analysis

A size-exclusion chromatography-organic carbon detection (LC–OCD) system (Modle 9, DOCLABOR, Germany) equipped with a chromatographic column (Toyopearl TSK HW50S, TOSOH Bioscience GmbH, Germany) was used to characterize DOM in the AnMBR, and a software program (Chrom CALC, DOC-LABOR, Germany) was used for data collection and analysis. Phosphate buffer solution (2.6 g/L KH_2PO_4 + 212.9 g/L Na_2HPO_4) with a pH value of 6.4 was used as the mobile phase at a flow rate of 2 mL/min, and the sampling time was set at 70 min. The dissolved organic carbon concentration of the total DOM and each fraction was measured, and the fraction of each sub-organic component was calculated as the ratio of its respective concentration to the total DOM.

2.5. Ex-situ filtration test and specific foulants resistance calculation

To determine the membrane resistance compositions, a filtration test was performed at the end of phases I and II, as reported previously (Chen et al., 2017b). Briefly, the membrane was cleaned by physical water scouring (to remove the cake layer), sponge swab (to remove the gel layer), 0.1% NaClO solution soaking (24 h, to remove the organics in membrane pores), and 10 g/L citric acid solution soaking (4 h, to remove the inorganics in membrane pores). After each cleaning step, a filtration test was performed in pure water three times. The resistances of the different components were calculated based on the value difference before and after each cleaning step. Based on the filtration resistance and foulant content (calculated using the total solids, TS) per membrane area in the different surface layers, the specific foulant resistance can be calculated by dividing the filtration resistance by the TS on the membrane.

2.6. Other analytical methods

The COD concentration was assayed using a rapid digestion-spectrophotometric method employing potassium dichromate as the oxidizer (APHA, 2005). Protein (PN) and polysaccharide (PS) concentrations were determined using the modified Folin Ciocalteu colorimetry method and phenol-vitriolic acid colorimetry method, respectively (Nielsen, 2010; Waterborg, 2002). The TS of the foulants and volatile suspended solids (MLVSS) in mixed liquor in the AnMBR were measured using weight difference analysis (APHA, 2005).

The percentage of CH_4 in the biogas was detected using a gas chromatograph (GC7900, Tianmei, Cina). For volatile fatty acids (VFAs) analysis, a mixed solution of 0.1 mol/L hydrochloric solution and a sample with a ratio of 1:1 (v/v) was injected into a gas chromatograph vial, and then the sample was assayed using a gas chromatograph (A91, PANNA, China). The particle size distribution was determined using a laser granularity distribution analyzer (LS 230/SVM, Beckman Coulter, USA) with a detection range of 0–2000 μm. The filtration property of the bulk sludge was determined by dead-end batch filtration tests according to the method of Ognier (Ognier et al., 2002). The electron transfer system activity of the bulk sludge was determined according to (Tian et al., 2017). The zeta potential of the mixed liquor was measured using a zeta potential meter (Zetasizer Nano ZS90, Malvern Instruments, UK).

The virgin and used biochar at the end of phase II were collected and air-dried, their specific surface area was measured by Brunauere-Emmette-Teller analysis separately, as reported in a previous study (Lei et al., 2019). The functional groups of the virgin and used biochar were identified using attenuated total reflectance-Fourier-transform infrared spectroscopy (ATR-FTIR) (IS50 FT-IR, Thermo Scientific Nicolet, UAS) with wave numbers in the range of 500–4000 cm^{-1}. The morphology of the virgin and used biochar was observed using a scanning electron microscope (SEM; MLA650F, FEI, USA).

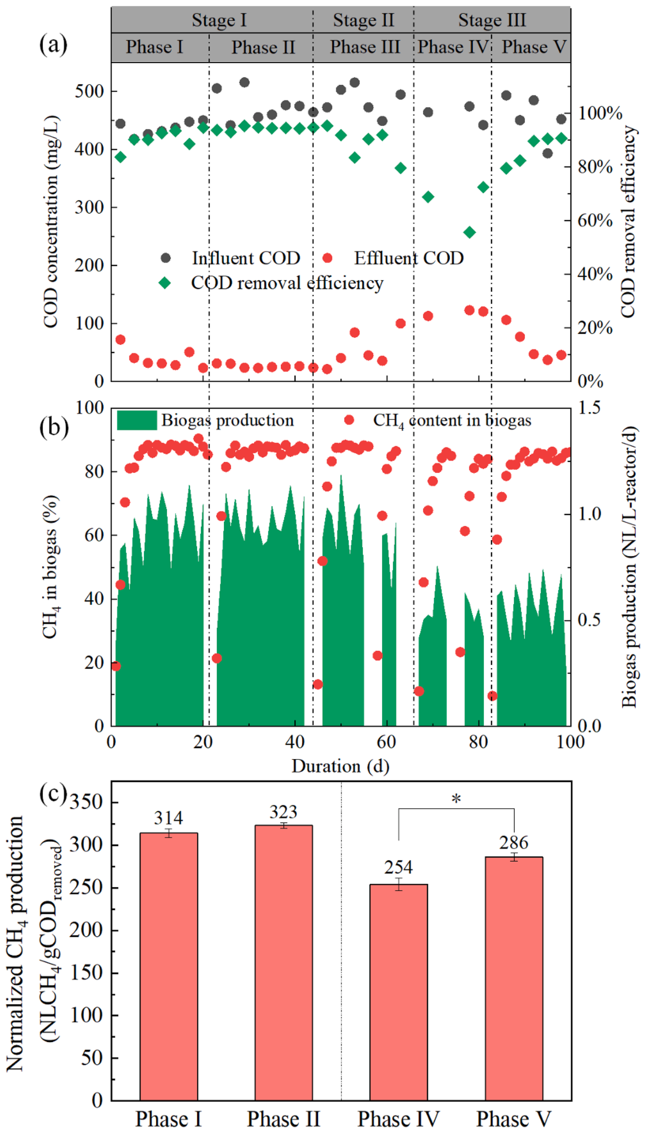

Fig. 2. Effects of biochar on AnMBR performance. (a) COD removal, (b) Biogas production and CH_4 content in biogas, and (c) normalized CH_4 production. "*" in figure (c) means these two groups are significantly different.

3. Results and discussion

3.1. Digestion performance

The AnMBR was operated for over 100 days in this study at a short HRT of 4.0 ± 0.3 h. The effluent COD was<50 mg/L in phases I and II, and a COD removal efficiency of over 85% was achieved, indicating that AnMBRs are efficient for COD removal at 18 ± 0.5 °C (Fig. 2a). Effluent COD increased rapidly when the temperature declined to 10 ± 0.5 °C (phase III, Fig. 2a), causing the COD removal efficiency to decrease to<60% (phase IV), mainly because of the presence of VFAs (mainly propionic acid and acetic acid) in the effluent (Table 1). Biochar addition improved the AnMBR performance, and the COD removal efficiency recovered to an average value of 86.6% (phase V). Digestion performance analysis showed that, although temperature showed a feasible impact on the CH_4 content in biogas, biogas production was largely reduced when the temperature decreased from 18 to 10 °C (Fig. 2b).

A surprising normalized CH_4 production of over 310 NmL/gCOD$_{removed}$ was achieved in phase I (theoretical value is 350 NmL/gCOD$_{removed}$), while it declined to only 254 ± 7 NmL/gCOD$_{removed}$ when the temperature declined to 10 °C in phase IV (Fig. 2c). Consistent with the COD removal efficiency, biochar addition largely increased CH_4 production, especially at lower temperatures. COD balance analysis revealed that CH_4 in biogas accounted for over 70% of the influent COD, while it decreased to<44% in phase IV (Table 2). The sludge growth only accounted for 5–8% of the influent COD at 18 °C, and it decreased to 2–5% of the influent COD at 10 °C, which is significantly lower than the value of 12% reported in our previous study (Lei et al., 2020), demonstrating that low temperature decreases sludge growth. Biochar addition had a positive impact on both methane production and sludge growth. CH_4 production increased by over 15% when biochar was added, suggesting that biochar can significantly enhance the energy recovery potential of AnMBRs. The electron transfer system activity was stimulated with the increased value from 3.2 ± 0.3 to 4.7 ± 0.1, which may be caused by the enhanced syntrophic VFAs oxidation by the addition of biochar (Table 3).

Table 2
COD balance of the AnMBR at different operating stages.

Operating stages	CH_4 in biogas	Dissolved CH_4 in effluent	Sludge growth	Effluent COD	Others
Phase I	73.9%	9.3%	5.3%	7.2%	4.3%
Phase II	77.5%	9.4%	7.5%	5.7%	0%
Phase IV	43.9%	11.5%	2.1%	23.6%	18.8%
Phase V	58.8%	11.9%	4.5%	13.6%	11.2%

Table 3
Properties of mixed liquor at different operating conditions.

Operating stages	Filtration property (/m/kg)	Zeta potential (mV)	Average diameter (μm)	Electron transfer system activity (μg/mg/h)	MLVSS (g/L)
Phase I	(10.6 ± 0.4) × 10^{16}	-(24.9 ± 1.9)	24.5 ± 0.7	/	8.5 ± 1.0
Phase II	(4.0 ± 1.5) × 10^{16}	-(25.5 ± 0.3)	19.1 ± 0.1	/	
Phase IV	(15.1 ± 2.1) × 10^{16}	-(27.6 ± 0.2)	19.8 ± 0.5	3.2 ± 0.3	
Phase V	(3.5 ± 0.9) × 10^{16}	-(27.9 ± 0.2)	24.0 ± 1.0	4.7 ± 0.1	

"/" means data are not collected.

3.2. Impacts of biochar addition on properties of mixed liquor

DOM concentration and sludge properties are the main indices of the mixed liquor that are closely related to AnMBR performance. In this study, the sludge concentration was maintained at a constant MLVSS value of 8.5 ± 1.0 g/L to exclude the impact of sludge concentration on the AnMBR performance (Table 3). DOM in the mixed liquor showed a higher concentration in stage III than in stage I (Fig. 3a), which could be attributed to the presence of VFAs at low temperatures. In addition, PN and PS concentrations also increased significantly after biochar was added ($P < 0.05$), which may be because electron transfer system activity was enhanced by biochar addition, hence causing an increase in PN and PS as metabolites. Although the zeta potential and particle size distribution (average diameter) showed no significant variation with biochar addition, the filtration property of the mixed liquor shows that the specific filtration resistance of the bulk sludge decreases significantly with the addition of biochar ($P < 0.01$) (Table 3), meaning that DOM may play a core role in causing the increase of specific filtration resistance.

Through LC-OCD analysis, six fractions, including hydrophobic organic carbon, biopolymers (BP, greater than 20 kDa), humic substances (HS, ~1 kDa), building blocks (BB, 300–500 Da), low molecular weight neutrals (LMWN, <350 Da), and low molecular weight acids

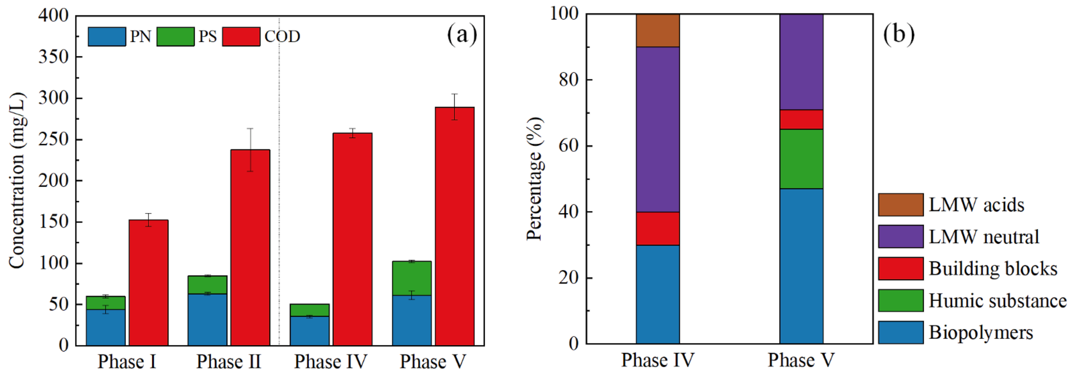

Fig. 3. Impacts of biochar on (a) DOM concentration and (b) composition (stage III).

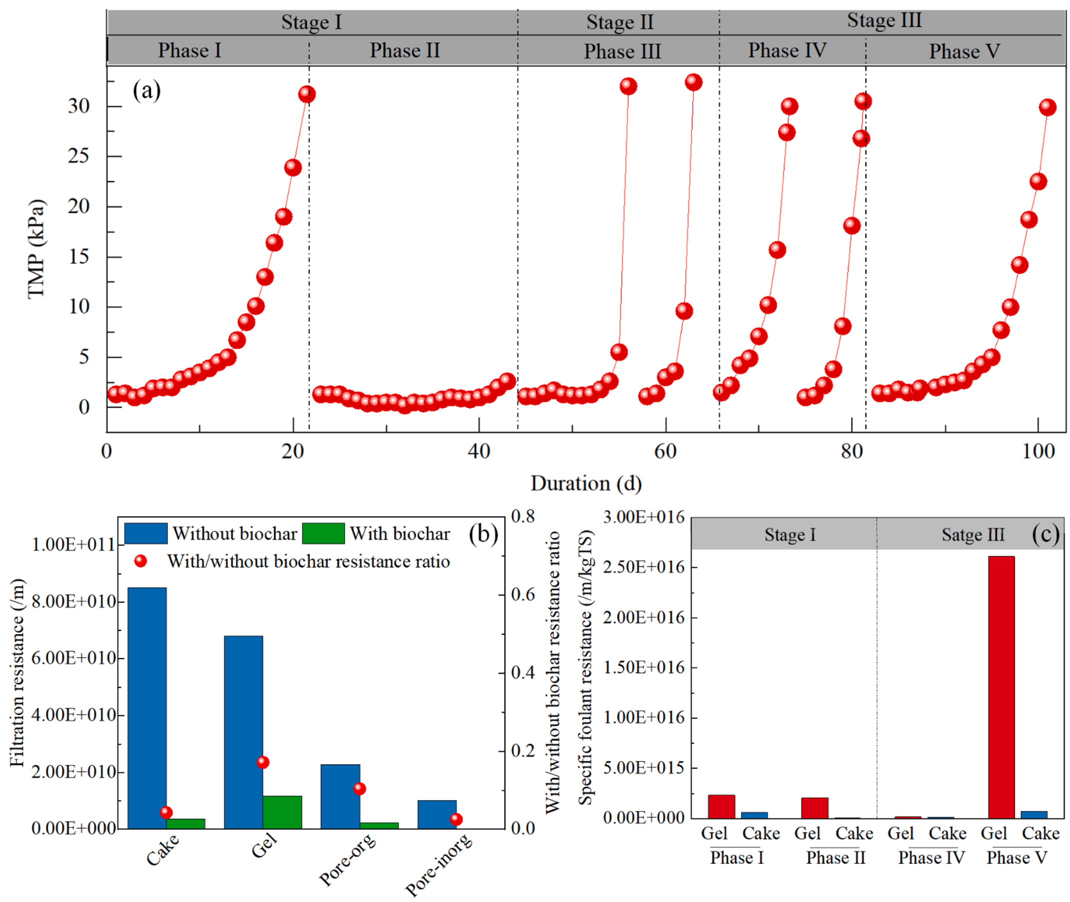

Fig. 4. Effects of biochar on membrane fouling performance. (a) TMP profile; (b) membrane resistance distribution (stage I); (c) specific foulants resistance.

(LMWA, < 350 Da) in DOM were analyzed (Fig. 3b). Hydrophobic organic carbon is organic matter that remains on the column caused by the strong hydrophobic interaction, which accounted for 89% of the dissolved organic carbon in phase IV, while it was undetected in phase V, indicating that biochar addition improved the hydrophilic property of DOM, which may be mainly attributed to the adsorption of biochar to hydrophobic organics in DOM. Compared with phase IV, the proportion of BP in phase V was significantly higher (30% *vs.* 47%), which may be because biochar addition promoted microbial metabolism in the AnMBR (Fig. 3b). LMWN comprises alcohols, aldehydes, ketones, sugars, and amino acids, which are the degradation products of BP (Laksono et al., 2020), and showed a higher proportion in phase IV than in phase V. This type of LMW substance can easily pass through membrane pores, so their high content in the DOM of this phase should be attributed to the low uptake rate of microbes to these substances.

3.3. Effects of biochar addition on membrane fouling performance

The TMP profile and membrane resistance were used to evaluate the effects of biochar addition on membrane performance (Fig. 4). In stage I (18 °C), the AnMBR was operated at a high average flux of 11.0 L/m^2/h for 21 days before TMP reached 30 kPa (phase I); in comparison, the TMP of the AnMBR in phase II was only 2.6 kPa after the same duration as phase I, showing the retarding effect of biochar in preventing membrane fouling (Fig. 4a). In stage II, the TMP showed a higher increase rate than that in stage I, and this situation was aggravated along with the operation duration. When the operating temperature declined to 10 °C (stage III), the AnMBR could be stably operated for only 7 days at an

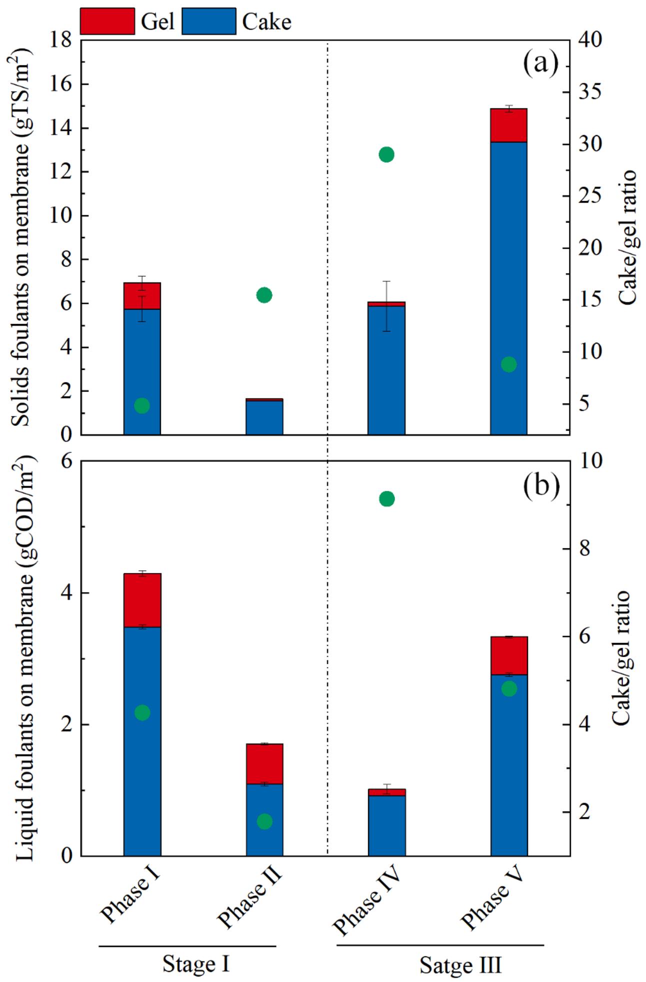

Fig. 5. Effects of biochar on (a) solid and (b) liquid foulants.

average membrane flux of 6.5 L/m^2/h (phase IV); the operation duration of the AnMBR was prolonged by 2.5 times in phase V when biochar was added, demonstrating that biochar can be used under different operating conditions. The membrane resistance composition analysis showed that the foulant resistance was ranked in the order of cake > gel > pore-org > pore-inorg (Fig. 4b). Among these, cake and gel layers accounted for over 75% of the total resistance (see Supplementary Material), indicating that they are the main foulants causing membrane fouling. The fouling resistance ratio with/without biochar was used to evaluate the impact of biochar on different foulants. The ratio of each component is much lower than 1.0, which is consistent with the TMP. However, the fouling resistance ratio with/without biochar of cake layer foulants is<0.1, which is much lower than the ratio of gel layer foulants (approximately 0.2), demonstrating that biochar is more effective in mitigating cake fouling. The specific foulant resistance of different surface layers revealed that the gel foulant resistance of the AnMBR with and without biochar was comparable (18 °C), while the AnMBR with biochar (phase II) has a lower cake foulant resistance, mainly because the cake layer of the AnMBR in this phase was not formed at a low TMP. Comparing the two fouled membranes in stage III (Fig. 4c), the specific foulant resistance of both the cake and gel layers with biochar (phase V) is much higher than that of without biochar (Phase IV). According to previous studies (Aslam et al., 2017; Wang et al., 2018c), adding carriers will enhance the scouring intensity of mixed liquor at the membrane surface. At this condition, foulants with large size will be shed from the membrane surface at a higher back-transport rate, which results in a compact fouling layer on the membrane surface, thereby increasing the specific foulant resistance.

To further evaluate the effects of biochar addition on cake and gel fouling, the foulants mass (evaluated using TS) and the compositions of the cake and gel foulants were investigated (Fig. 5). For the AnMBR operated for the same duration (phase I and phase II), the solid foulants in both the cake and gel layers in phase I were much lower than those in phase II (Fig. 5a), indicating that biochar retarded the accumulation of solid foulants in both the cake and gel layers. For the fouled membrane (TMP = 30 kPa, 10 °C), more foulants accumulated in both the cake and gel layers in phase V than in phase IV, this maybe because the distribution of foulants structure was changed. Moreover, the cake/gel ratio decrease from 29.0 to 8.8 when biochar was added, suggesting that biochar is more effective in retarding solid foulants in the cake layer. Similarly, liquid foulants in the cake layer in phase IV accounted for only 44% of the AnMBR in phase V, whereas in the gel layer it was comparative (0.82 gCOD/m^2 *vs.* 0.61 gCOD/m^2) (Fig. 5b), indicating that biochar is more effective in retarding liquid foulants in the cake layer.

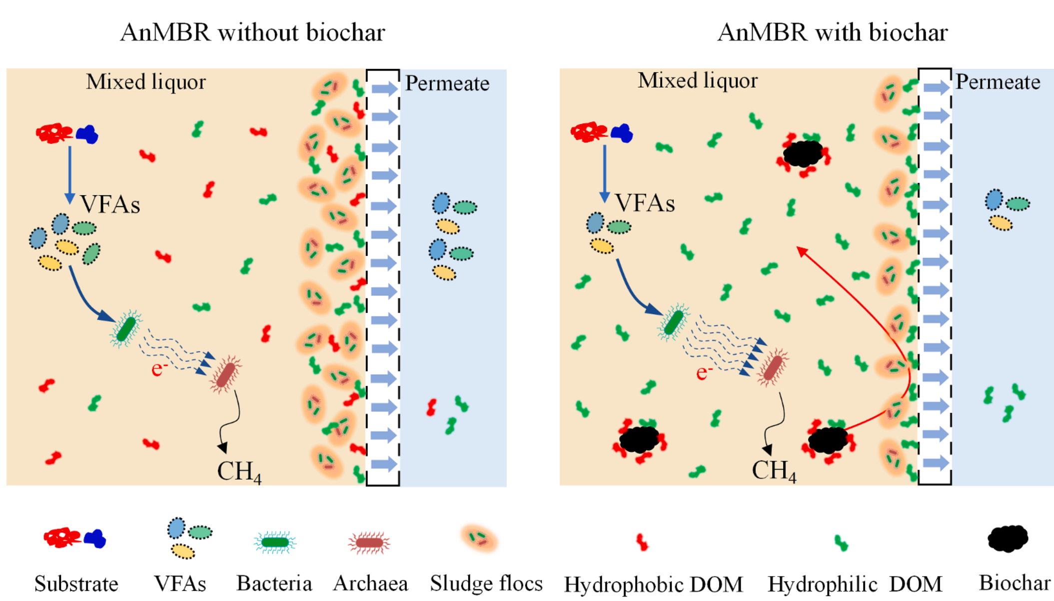

Fig. 6. Potential roles of biochar in digestion enhancement and fouling control.

3.4. Potential roles of biochar in digestion enhancement and fouling control

According to the SEM images of the virgin and used biochar, both biopolymers and microbes were observed attached to the biochar (see Supplementary Material), while nearly no biomass can be observed in the inner pores of biochar. Although the specific surface area of biochar decreased from 37.8 ± 0.2 to 0.33 ± 0.13 m^2/g after used, the volatile solids on biochar were only approximately 0.035 ± 0.007 g VSS/g biochar, meaning that<1% of the total volatile solids in the bulk sludge were absorbed on biochar. The low attached biomass quantity and the significantly decreased specific surface area of biochar may be caused by the covering of polymers and microbes on the surface pores of biochar. Four main peaks, including O–H stretching (3427 cm^{-1}), asymmetric/symmetric C–H stretching (2924 cm^{-1}), C=O stretching (1572 cm^{-1}), and asymmetric or symmetric C–O stretching or C–O deformation (1048–1200 cm^{-1}) were identified in both virgin and used biochar (see Supplementary Material). The presence of these functional groups is benefited from the low pyrolysis temperature of biochar and is believed to play key roles in promoting digestion efficiency (Wang et al., 2018a). The oxygen-containing group (including C=O and C–O), especially C–O, showed a significant decrease in the used biochar, which is consistent with a previous study (Rechberger et al., 2017), this may be because they acted as a redox agent in anaerobic digestion.

The potential roles of biochar in DOM generation and fouling reduction were proposed based on findings from the present study (Fig. 6). Biochar has been reported to promote VFAs utilization through syntrophic oxidation and electron transfer (Li et al., 2018; Wang et al., 2019), in which, the oxygen-containing group in biochar promotes electron transfer between electroactive microbes (Wang et al., 2020), this should be the main mechanism that biochar promotes digestion in the current study. As for membrane fouling control, the enhanced scouring intensity of the mixed liquor at the membrane by granular additives has been reported elsewhere (Wang et al., 2017; Zhang et al., 2021). At a higher scouring intensity, surface fouling was largely restarted, especially the cake layer, which resulted in a relatively severe gel fouling. Furthermore, biochar is hydrophobic, which facilitates the adsorption of biochar to hydrophobic organics that have higher fouling potential in DOM and hence facilitates membrane fouling control. The strategy of optimizing AnMBR operation at low temperatures using biochar as an additive is economically feasible and environmentally friendly. At a low adding dosage, biochar showed comparable effects to digestion performance and membrane fouling control with activated carbon reported previously (Hu and Stuckey, 2007), meaning that it has a lower investment than activated carbon because biochar needn't be activated. In addition, granular biochar has advantages to avoid the washout of biochar during sludge discharge and the increase of sludge yield than powdered additives (Remy et al., 2009), suggesting this strategy has a higher potential to be applied in the real case.

4. Conclusions

Biochar supported high digestion performance and low membrane fouling rate in AnMBRs treating sewage at low temperatures. COD removal and methane recovery were largely enhanced, in which VFAs utilization was promoted by the extracellular electron transfer system activity with the assistant of biochar. Biochar retarded the membrane fouling by reducing the hydrophobic DOM in AnMBR and accumulation of large particles on the membrane surface, which decreases the cake fouling, alters fouling layer compositions and filtration property. Thus, biochar (and the underlying action mechanisms) can serve as a practical approach for improving AnMBR performance.

CRediT authorship contribution statement

Zhen Lei: Data curation, Formal analysis, Writing - original draft. **Yu Ma:** Investigation, Data curation. **Jun Wang:** Investigation, Data curation. **Xiaochang C. Wang:** Conceptualization, Supervision. **Qian Li:** Project administration. **Rong Chen:** Project administration, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biortech.2021.124966.

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