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Sludge aggregation and biofouling mitigation in A/O-MBR treating anaerobically digested swine slurry with PAC/PAM flocculation pretreatment

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ABSTRACT

Sludge aggregation and biofouling mitigation in anoxic/oxic membrane bioreactor (A/O-MBR) treating anaerobically digested swine slurry with PAC/PAM flocculation pretreatment were studied in this study. Two laboratory A/O-MBRs were set-up and operated with anaerobically digested leachate with and without PAC/PAM flocculation pretreatment, respectively, for over 150 days. Zeta potential, average size, soluble microbial products (SMP), extracellular polymeric substances (EPS), microscope, relative hydrophobicity (RH), etc. of sludge characteristics were detected in this study. Flocculation pretreatment with 1600 mg PAC/L wastewater and 50 mg PAM/L wastewater effectively optimized the anaerobically digested swine slurry with the obvious COD_{Cr} and TP removal for further biological treatment. Meanwhile, PAC/PAM pretreatment caused approximate 55–74 mg/L Al flowing into A/O-MBR, leading to sludge aggregation and high density of biomass. PAC/PAM pretreatment also caused SMP decrease and RH increase, and bridged polysaccharides to enhance gelation. EPS decrease further contributed to the decrease of the membrane mass transfer coefficient. PAC/PAM flocculation pretreatment effectively led to the sludge aggregation and further caused biofouling mitigation.

Keywords: Sludge aggregation; PAC/PAM flocculation; Biofouling mitigation; Anaerobically digested swine wastewater

1. Introduction

With population growth, the increasing worldwide need for aliment has caused the intensification of livestock production in the recent three decades. Especially in China, the swine husbandry is the most important agricultural industry in the rural areas of China, and the stock of swine are over 500 million [1]. However, the swine wastewater, containing high concentration of organic components and inorganic nutrients (such as COD_{Cr} (chemical oxygen demand), NH_4^+–N, total nitrogen (TN), total phosphorus (TP), Ca^{2+}, etc.), has been considered as the major pollution originating from swine husbandry, and over 6.0 billion tons swine wastewater is annually generated from the swine waste in China, which caused the severe harm to the environment [2,3]. Consequently, many countries are focused on the swine wastewater treatment due to tightening legislation and standards.

Generally, swine wastewater is treated in three methods of land spreading, natural treatment, and engineering treatment [4,5]. But, land spreading and natural treatment need large land area, which is the obstacle for most of land-restricted swine farm, thus engineering treatment is the desirable choice for swine husbandry. Anaerobic digestion is often regarded as the alternative for swine wastewater treatment due to its high-effective organic matter removal, methane generation, pathogen stabilization, odor reduction, energy recovery, etc. [6]. However, large volume of anaerobically digested swine slurry (known as "digested piggery wastewater" or "swine wastewater digest-ate") still contains high concentrations of COD, NH_4^+–N, TN, Ca^{2+}, Zn^{2+}, Mg^{2+}, bioactive substances (such as protein, amino acids, in-dole acetic), growth hormone, etc. It would be

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difficult issue for the spreading of all effluent on land due to those pollutants [1]. Therefore, anaerobically digested swine slurry is considered as one of unwieldy wastewater in environmental engineering area [7–9]. Additionally, anaerobically digested swine slurry is also regarded as one of recyclable source for an effective fertilizer due to its rich nutrients in recent years.

As the technology combining physical separation and biological degradation, membrane bioreactor is the high-efficiency treatment for industrial, municipal and livestock wastewater [10–12]. Compared with traditional biological wastewater treatment, MBR presents outstanding advantages of better sludge regulation, low footprint, less sludge production, high quality of effluent, etc., and thus its application has been widely promoted all over the world [13]. Furthermore, due to its outstanding advantages, MBR is prior to being applied for anaerobically digested swine slurry treatment, especially for the high concentration of ammonia removal, antibiotic and hormones elimination, etc. [14–16]. Prado et al. [17] has applied semi-industrial MBR for effective swine wastewater treatment. Additionally, dozens of large-scale MBR over 10,000 m^3/d wastewater treatment have been built-up and operated only in China, and the total capacity of large-scale MBR in China has exceeded 11.17 $\times 10^6 m^3/d$ by the end of 2017 [18,19]. Consequently, MBR has been already considered as a mature technology for engineering application. In addition, MBR also applied as the combining technology for nitrogen and phosphorus recovery. Our previous works (Fig. S1; National Science and Technology Pillar Program: 2013BAD21B03) had built up a recovery system for swine wastewater from an over 3000 pigs farm, and this system aimed to recycle nutrients from swine wastewater as the effective fertilize to grow grass. Swine wastewater was first anaerobic digested with up-flow anaerobic sludge bed (UASB), then anaerobically digested swine slurry was treated with anoxic/oxic membrane bioreactor (A/O-MBR). The effluent of A/O-MBR was purified as rich nutrients (known as fertilizer) and applied for grass growth after advanced treatment. However, plenty of inorganic and organic particles flew into A/O-MBR with anaerobically digested swine slurry during the recovery system operation, which would lead to low pollutant removal, severe membrane biofouling and high running-cost for purification. Engineering economic significance of this recovery system is challenged. As previous studies reported [20–22], flocculant addition has been reported as one of the most common solutions for the improvement in effluent water quality, especially the removal of organic components and phosphorus (adjusting the ratio of each components in swine wastewater) and thus flocculation could effectively optimize anaerobically digested swine slurry for biological treatment. $FeCl_3$, $Al_2(SO_4)_3$, polyaluminum chloride (PAC), polyacrylamides (PAM), Ca^{2+}, Mg^{2+}, bioflocculant, etc. have been applied for wastewater treatment, and previous literatures [23,24] have reported that PAC was one of best flocculant for swine wastewater treatment. Normally, for organic components, cations induced by PAC serve as flocculant and facilitate flocculation with the result of double electrical layers and ion-bridge through extracellular polymeric substances (EPS). Moreover, the continuous compression of double electrical layer still leads to further biological flocculation improvement due to high concentration of cation [25]. In addition, aluminum salt would improve the removal inorganic phosphorus. Compared with other aluminum salt, PAC provides stronger and faster settling flocs due to its polymerization process, and PAC does not decrease pH as much as other aluminum salts [23]. PAM is also applied with PAC for better flocculation during wastewater treatment. PAC and PAM are the common commercial products and suitable for the wide application. Consequently, PAC/PAM flocculation was applied for slurry pretreatment in the laboratory scale before engineering application, and we figured out both sludge aggregation and membrane biofouling mitigation during A/O-MBR operation with flocculation pretreatment. However, previous studies are mainly focusing on swine wastewater treatment with pretreatment, but anaerobically digested swine slurry treatment is generally less studied [26,27]. Thus, performance of PAC/PAM pretreatment for anaerobically digested swine slurry treatment is still need the further study, especially sludge aggregation and biofouling mitigation.

This study aimed to figure out performance of PAC/PAM pretreatment for anaerobically digested swine slurry treatment, and especially identified effects of sludge aggregation on biofouling mitigation. Two laboratory-scale A/O-MBRs were operated with anaerobically digested leachate with and without PAC/PAM flocculation pretreatment, respectively, for over 150 d. Sludge characteristics were detailed with *Zeta* potential, average size, soluble microbial products (SMP), EPS, microscope, relative hydrophobicity (RH), etc., to analyze the interaction between sludge aggregation and biofouling mitigation.

2. Materials and methods

2.1. Set-up and operation of reactors

Two laboratory-scale A/O-MBRs, containing 2.7 L anoxic tank and 4.5 L oxic tank (similar as anoxic:oxic tank ratio as our practical scale A/O-MBR), were operated with and without PAC/PAM flocculation pretreatment (MBR-PP and MBR-Control), respectively. A polyvinylidene fluoride (PVDF) hollow fiber membrane module (0.04 μm of pore size; 0.02 m^2 of total surface area; Litree Company, China) was set in the oxic tank of each MBR, and air scouring system installed at bottom membrane module. Constant fluid flux (7.5 mL/h) was operated in an intermittent suction cycle of 8 min on/ 2 min off. Because anaerobically digested swine contained low biodegradable substances and longer reaction time was needed, hydraulic retention time (HRT) and solids retention time (SRT) were both maintained at 50 days (HRT of anoxic tank was 50 days). HRT in this study was longer than normal HRT, which was because anaerobic digested swine slurry contained toxicity components and short HRT would increase toxicity exposure to MBR. Moreover, lab-scale MBR had lower shock tolerance than practical scale MBR, and lab-scale MBR had a higher HRT during operation. Flow rate of recycled mixed liquor from oxic tank to anoxic tank was controlled at 400% of the influent flow rate. Temperature and pH of each MBR was remained in the range of 25–29°C and 7–8 (with NaOH solution, normally maintained the influent (anaerobically digested swine slurry after PAC/PAM pretreatment) at 7.2–7.5), respectively. Dissolve oxygen (DO) of anoxic and oxic tank were controlled at <0.5 mg/L and 2.0–4.0 mg/L, respectively.

Anaerobically digested swine slurry was the effluent of an up-flow anaerobic sludge bed (UASB) for fresh piggery wastewater treatment, which was from a swine farm containing over 3000 pigs (Suzhou, Jiangsu Province, China). The original anaerobically digested swine slurry was characterized in Table S1 (Supporting Information). During this study, the swine farm was operated with the approximately similar conditions, indicating that anaerobically digested swine slurry was relatively stable in this study.

Return activated sludge stream in the Quyang wastewater treatment plant (Shanghai, China) was applied as the inoculating sludge in this study. Inoculating sludge was initially cultured with the mixture of municipal wastewater and anaerobically digested swine slurry, then gradual increase of slurry was performed to promote bacteria adapting to operational conditions of MBR-PP and MBR-Control, respectively. When the treatment performance of reactor was stable, membrane module was replaced with a new unit and A/O-MBR was operated for over 150 days for this study. When transmembrane pressure (TMP) reached 40 kPa, the membrane module was removed with backwashing, physical (washing with tap water) and chemical cleaning (1% NaOCl and 10% citric acid immersion for 6 h, respectively) to recover the membrane permeability. Moreover, 2 M NaOH and 2 M HCl were applied in this study for pH adjustment.

2.2. Optimum batch experiment for PAC/PAM flocculation pretreatment

Optimum batch experiment was carried out to estimate the optimal condition of PAC and PAM to remove parts of pollutants in anaerobically digested swine slurry. The dosage of PAC and PAM was selected 400–3200 and 50–200 mg/L, respectively, according to pre-experiments, and then further analyze the operational concentration of PAC and PAM in this optimum batch experiment. Anaerobically digested swine slurry was performed in 500 mL beaker with the addition of certain PAC and PAM. Slurry was first mixed with 400 rpm for 2 min, then blended at 150 rpm for 15 min. After 1.5 h sedimentation (similar as the PAC/PAM flocculation tower operation), the supernatant was analyzed for the removal efficiencies of COD_{Cr}, TP and NH_4^+–N.

2.3. Batch experiment for *Zeta* potential and average size of sludge flocs with PAC

Batch experiment was carried out to identify the effects of PAC on *Zeta* potential and average size of sludge flocs. Moreover, the influent of A/O-MBR (anaerobic digested swine slurry after PAC/PAM pretreatment) was approximate 55–74 mg Al/L, and thus concentrations of 0, 10, 20, 40, 60 and 80 mg Al/L were carried out for better understanding the effects of PAC on *Zeta* potential and average size of sludge flocs. 25 mL sludge from MBR-Control was performed in 50 mL beaker with 0, 10, 20, 40, 60 and 80 mg Al/L PAC. Sludge was mixed with 200 rpm for 10 min. Then the mixed liquor of sample was first shaken to break flocs into small particles, and the supernatant was sampled for *Zeta* potential with *Zeta* sizer Nano Z (Malvern Instruments Ltd., UK). In addition, average size of sludge flocs in mixed liquor was directly measured with a focused beam reflectance measurement (Eyetech particle size and shape analyzer, Ankersmid, Holland).

2.4. Extraction and measurement of SMP and EPS

SMP and EPS from sludge were extracted based on a modified thermal extraction method [28,29]. 40 ml sludge was 5-min centrifuged (MILTIFUGE X1R, Thermo Electron Corporation, USA) at 6000 g, and the filtered supernatant (with 0.45-μm filter (SCAA-101, ANPEL, China)) was considered as SMP. Then remaining sludge was re-suspended with 40 ml 0.9% NaCl solution, and shaken at 150 rpm for 10 min after 15 min ultrasound treatment (DS510DT, 40 kHz, 300 W, Shangchao, China). The sludge was further heated at 80ºC for 30 min. Next, sludge was centrifuged at 12000 g for 20 min, and the supernatant was regarded as EPS. SMP and EPS were normalized as the concentration of polysaccharide, protein, and total organic components (represented as TOC). They were measured by the phenol-sulfuric acid method, Branford method, diphenylamine method and TOC analyzer (TOC-V_{VPN}, Shimadzu, Japan), respectively. Molecular weight (MW) distribution was determined with a gel filtration chromatography (GFC) analyzer, consisting of a TSK G4000SW type gel column (TOSOH Corporation, Japan) and a liquid chromatography spectrometer (LC-10ATVP, SHIMADZU, Japan). Samples were also applied for three-dimensional excitation-emission matrix (EEM) fluorescence spectroscopy analysis (detailed in SI).

2.5. Membrane resistance analysis

Total membrane resistance was classified into fresh membrane resistance, pore blocking resistance, concentration polarization resistance and cake layer resistance. Resistance was calculated with TMP, permeate flux and viscosity according to Zhou et al. [30]. Analysis was detailed in SI.

2.6. Others analysis

The standard methods were used to measure concentrations of NH_4^+–N, COD, TN, TP, turbidity, mixed liquor suspended solid (MLSS) and mixed liquor volatile suspended solids (MLVSS) as well as the sludge volume index (SVI) [31]. A focused beam reflectance measurement (Eyetech particle size and shape analyzer, Ankersmid, Holland) was used to identify the particle size of sludge. Sludge with 20-time dilution was also detected through CX22 microscope (Olympus Corporation, Japan). DO and pH were measured with a DO-and-pH meter (HQ4d, HACH, USA). RH was evaluated similar to Meng et al. [32] (Detailed in SI). Inorganic elements in the influent and effluent were detected with an inductively coupled plasma-optical emission spectrometer according to the Standard Methods [33]. Oxygen uptake rate (OUR) was measured based on Zhou et al. [34]. The concentrations of aluminum in each fraction were determined by ICP-OES (Optima 2100 DV, Perkin Elmer, USA) after HNO_3 digestion.

3. Results and discussion

3.1. PAC/PAM flocculation pretreatment

Swine slurry is one of awkward livestock wastewater, containing high concentrations of COD_{Cr}, NH_4^+-N, TP, volatile organic compounds (VOCs), humic acid substances, antibiotic, etc. [35,36]. Especially, anaerobically digested swine slurry has the low C/N rate with plenty of non-biodegradable COD_{Cr} after anaerobic digestion. Additionally, non-biodegradable COD_{Cr} and most of PO_4^{3-} cannot be biodegraded with advance biological treatment. Thus, solid-liquid separation is regarded as one of most effective application for the pretreatment of anaerobically digested swine slurry [22]. In previous literatures [21,22,37], Fe^{3+}, Al^{3+}, PAM, bioflocculant, etc. have been applied, respectively, for the solid-liquid separation of fresh or anaerobically digested swine slurry. In addition, compared with other traditional activated sludge process, A/O-MBR can achieve high effective NH_4^+–N removal due to its excessive oxygen supply. Pretreatment should be focus on the removal of COD_{Cr} and TP. Optimum batch experiment (Table 1) was carried out to estimate the optimal condition of PAC and PAM for anaerobically digested swine slurry. Based on pre-experiments, PAC and PAM were selected the range of 400–3200 and 50–200 mg/L, respectively. Batch 1–6 shows that PAC addition could effectively increase the removal of COD_{Cr} and TP with similar PAM dosage. However, TP removal efficiency decreased with PAM addition (Batch 7–10). In addition, considering operational cost of treatment system, combination of PAC and PAM is the best choice for the solid-liquid separation of anaerobically digested swine slurry. Results of optimum batch experiments show that TP removal efficiency was enhanced with PAC increase but PAM decrease. It was because aluminum in PAC can effectively precipitate with PO_4^{3-} to reduce TP in the anaerobically digested swine slurry. Meanwhile regarding COD_{Cr} removal, positive charge PAM was applied in this study, but leading to the decrease of TP removal efficiency. COD_{Cr} was also removed due to the aggregation of organic components, which was because high concentration of cations, induced by PAC, led to the compression of double electrical layer and caused ion-bridging further resulting in organic components precipitate. In this project, the effluent of A/O-MBR was applied for applied for nutrients recovery from swine wastewater as the

Table 1
Optimum batch experiment of PAC/PAM flocculation pretreatment[a]

	PAC	PAM	TP	TP removal efficiency	COD_{Cr}	COD_{Cr} removal efficiency	NH_4^+–N	NH_4^+–N removal efficiency
Original			58.3		759		1101	
1	400	100	35.0	40.0	691	9.0	1061	3.6
2	800	100	33.0	43.4	552	27.3	1073	2.5
3	1200	100	26.8	54.1	390	48.6	1076	2.3
4	1600	100	23.5	59.6	408	46.2	1084	1.5
5	2000	100	19.3	66.9	414	45.5	1084	1.5
6	2400	100	19.1	67.3	430	43.3	1082	1.5
7	2000	50	15.6	73.3	395	47.9	1062	3.8
8	2000	100	17.2	70.6	392	48.3	1052	4.9
9	2000	150	17.3	70.4	467	38.4	1052	4.9
10	2000	200	19.2	67.2	397	47.6	1059	4.1
11	800	50	25.1	57.1	402	47	1063	3.4
12	1200	50	20.9	64.2	287	62.1	1060	3.7
13	1600	50	16.7	71.3	300	60.5	1049	4.7
14	2000	50	7.4	87.5	402	47	1063	3.4
15	3200	50	3.2	94.6	287	62.1	1060	3.7
16	1600	–	24.5	58.0	368	51.5	1064	3.4
17	–	50	47.4	18.7	720	5.1	1083	1.6

[a] Removal efficiencies of TP, COD_{Cr} and NH_4^+–N were %, and other items were mg/L.

effective fertilize to grow grass, and thus high TP removal of Batch 15 was not suitable for this project. Batch 16 and 17 further showed that co-application of both PAC and PAM had better pretreatment performance than using PAC or PAM alone. Consequently, the optimal condition for slurry pretreatment was selected as high as 1600 mg PAC/L, but chosen 50 mg PAM/L in this study.

3.2. *Sludge aggregation*

Table S2 showed the A/O-MBR performance with or without PAC/PAM pretreatment at optimal condition (1600 mg PAC/L and 50 mg PAM/L). MBR-PP would had better performance due to pollutant reduction with pretreatment. Moreover, A/O-MBR directly treating the raw swine wastewater also showed the poor degradation. Previous literatures pointed out that the combination with anaerobic, aerobic, physical and chemical methods need to be carried out for swine wastewater treatment due to the complex components in the wastewater, and thus single method could not effectively treat raw swine wastewater [15,17,38]. In addition, NH_4^+–N was mainly depended on A/O-MBR. However, although A/O-MBR had an excellent NH_4^+–N removal, anoxic tank did not present the effective TN degradation, because of low biodegradable carbon source. Anoxic tank in this project was not only for total nitrogen removal, but also to enhance the toxicity resistance of activated sludge, because anaerobic bacteria had higher toxicity resistance. After PAC (1600 mg/L)/PAM (50 mg/L) pretreatment, anaerobically digested swine slurry had over 70% TP and 60% COD_{Cr} removal, and also contained approximately 55–74 mg/L Al^{n+}, which flowed into A/O-MBR. This probably led to the sludge aggregation with aluminum flocculation. Consequently, batch experiments for *Zeta* potential and average size of sludge flocs with PAC was operated with 0, 10, 20, 40, 60 and 80 mg Al/L PAC. As Fig. 1 shows, MBR-PP had the obvious sludge aggregation and high density of biomass. Table 2 further presents that average size of sludge flocs in MBR-PP was 82 ± 8 µm, which was approximately twice of that in MBR-Control, indicating sludge aggregation. As previous literature [25] reported, high concentration of cations, induced by PAC, led to the compression of double electrical layer and caused ion-bridging through EPS and SMP, further promoting flocculation. Based on Derjaguin-Landau-Verwey-Overbeek (DLVO) theory, thinner double electrical layers tend to reduce the repulsive energy among sludge flocs. Therefore, sludge flocs had the effective aggregation with large size under PAC concentration advance. Both of Microscope images (Fig. 1) and average size (Table 2) of sludge showed obvious sludge aggregation and high density of biomass with PAC/PAM pretreatment. In addition, sludge floc normally is negative charge, and its mobility predicts the form of sludge flocs. The mobility of sludge floc in MBR-PP was only third of that in MBR-Control. PAC/PAM pretreatment induced *Zeta* potential decrease from –18.4 ± 1.3 to –7.2 ± 0.8 mV, which was similar as Wen et al. [25]. This was because of the double electrical layer theory: positive surface charging colloids, resulted from Al ion hydrolysis, would compress the double electrical layer, and thus lead to *Zeta* potential decrease [25]. Batch experiments (Fig. 2) were also carried out to identify the PAC effects on *Zeta* potential and average size of sludge flocs. With PAC concentration increase, *Zeta* potential of sludge flocs was obviously reduced, and sludge flocs had the effective aggregation with large size. Lower mobility of sludge flocs in MBR-PP indicated the neutralization of sludge floc surface charge and/or the increase of floc size. Additionally, results of *Zeta* potential further proved the neutralization of sludge floc, and indicated the instability of sludge colloid/floc and the tendency of sludge aggregation. Moreover, bridge effects of PAC also contributed the flocculation and coagulation for sludge aggregation, and Liu et al. [39] applied the bridge effects of PAC to granulate sludge flocs. Batch experiments (Fig. 2) showed that PAC could effectively induce sludge aggregation. Moreover, PAC/PAM pretreatment could effectively reduce the turbidity of anaerobically digested swine slurry from 190–220 to 110–120 NTU. The turbidity decrease of anaerobically digested swine slurry indicated that PAC/PAM pretreatment optimized the influent quality of A/O-MBR and intensified the sludge aggregation.

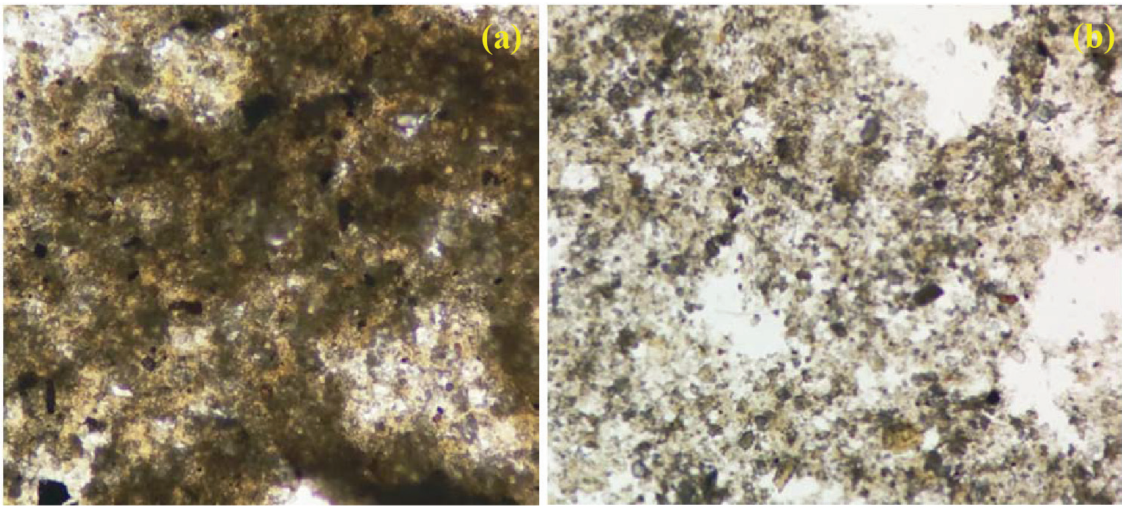

Fig. 1. Microscope image (100) of sludge in (a) MBR-PP and (b) MBR-control.

Table 2
Zeta potential, average size and mobility comparisons of sludge floc between MBR-PP and MBR-Control (n = 14)

	Zeta (mV) [a]	Average size (μm)	Mobility (μm cm/Vs)
MBR-PP	–7.2 ± 0.8	82 ± 8	–0.566 ± 0.038
MBR-Control	–18.4 ± 1.3	47 ± 5	–1.441 ± 0.143

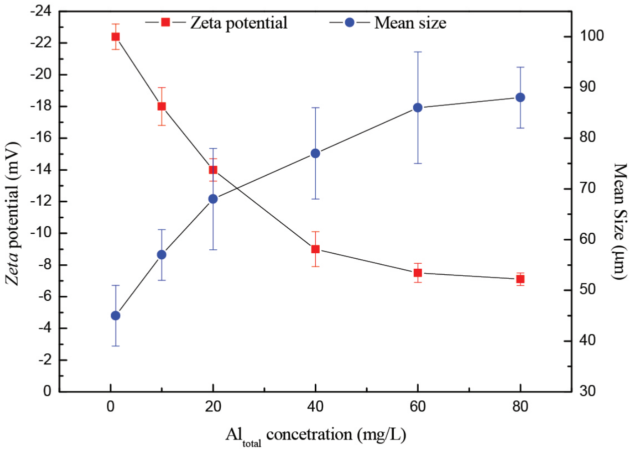

Fig. 2. *Zeta* potential and mean size variations of sludge floc in the batch experiment (n = 3).

3.3. Sludge characteristics

During 150 days operation, MBR-PP and MBR-Control maintained MLSS in the range of 6.4 ± 0.7 g/L and 9.6 ± 3.8 g/L, respectively, but remained the similar MLVSS at the range of approximate 4.5 ± 1.0 g/L. Similar MLVSS but various MLSS indicated plenty of the inorganic components accumulated in mixed liquor and PAC/PAM pretreatment effectively removed inorganic solid fraction in anaerobically digested swine slurry. Liu et al. [39] also presented the similar concentration of MLVSS in the physico chemical-biochemical treatment with PAC, and PAC could effectively increase the sludge flocs size. SVI is considered as the significant index of sludge, and good operational SVI is normally in the range of 50–100 mL/g [40,41]. SVI of MBR-PP was around 84 ± 10 mL/g, but MBR-Control SVI decreased gradually to 36.5 mL/g at the last 20 days. SVI variations between MBR-PP (84 ± 10 mL/g) and MBR-Control (43 ± 8 mL/g) indicated that anaerobically digested swine slurry without pretreatment reduced sludge viability, which was probably due to poor nutrient [40]. Additionally, OUR was carried out to identify the viability of sludge.

Table 3
Sludge characteristics of MBR-PP and MBR-Control (n = 14)

	MBR-PP	MBR-Control
MLSS (g/L)	6.4 ± 0.7	9.6 ± 3.8
MLVSS (g/L)	4.7 ± 0.8	4.2 ± 1.2
MLSS/MLVSS	73 ± 8	44 ± 12
SVI (mL/g)	84 ± 10	43 ± 8
OUR (mg/g MLSS·min)	0.0632 ± 0.0045	0.0485 ± 0.0073
RH (%)	65 ± 2	55 ± 3

[a] MLSS/MLVSS in anoxic tank: MBR-PP = 68 ± 7; MBR-Control = 42 ± 8.

Table 3 further shows that PAC/PAM pretreatment could effectively increase sludge viability from 0.0485 ± 0.0073 to 0.0632 ± 0.0045 mg/g MLSS·min. As the most frequently used additive in many animal feeds, cupric salts and antibiotics would inhibit the following biological treatment [42,43], and chemical precipitation is reported as one of effective method to remove the toxicity components and optimize the wastewater biodegradability [24,44].

As the significant characteristics of sludge on membrane biofouling, SMP and EPS are detailed in Table 4. PAC/PAM pretreatment led to the obvious decreases of polysaccharide, protein and TOC of both SMP and EPS. Fig. 3 presents the EEM spectra of SMP and EPS in both MBRs. The fluorescent peak A, at the Ex/Em of 340/420 nm, is related to visible humic acid-like substances [45], which is the common fluorescent component in anaerobically digested swine slurry [9,46]. Humic acid-like substances of SMP and EPS were also reduced with PAC/PAM pretreatment. However, PAC/PAM caused slightly MW decrease of SMP and EPS in sludge and only reduced the fraction >100 kPa (Fig. 4), which was similar as the result of Arabi et al. [47]. Previous literatures [47,48] reported that cation would decrease organic components rejection in membrane system, and further attribute to the reduction of repulsion between the organic components molecular. Thus, the high MW fraction was mainly affected by the cation, which was induced by PAC. In all, PAC/PAM pretreatment effectively reduced the concentration of SMP and EPS. Many studies [37,49–51] have reported that Al^{n+} and flocculants could remove SMP and EPS with sludge aggregation during flocculation and/or coagulation.

3.4. Membrane biofouling mitigation

TMP is the direct index reflecting the situation of membrane biofouling. TMP variations of MBR-PP and MBR-Control throughout the 150 days operation are

Table 4
SMP and EPS comparisons of sludge between MBR-PP and MBR-Control (n = 14)

	SMP			EPS		
	Polysaccharide	Protein	TOC	Polysaccharide	Protein	TOC
MBR-PP	78 ± 10	29 ± 5	321 ± 25	113 ± 14	26 ± 6	340 ± 35
MBR-Control	61 ± 6	11 ± 2	187 ± 15	64 ± 8	12 ± 2	215 ± 18

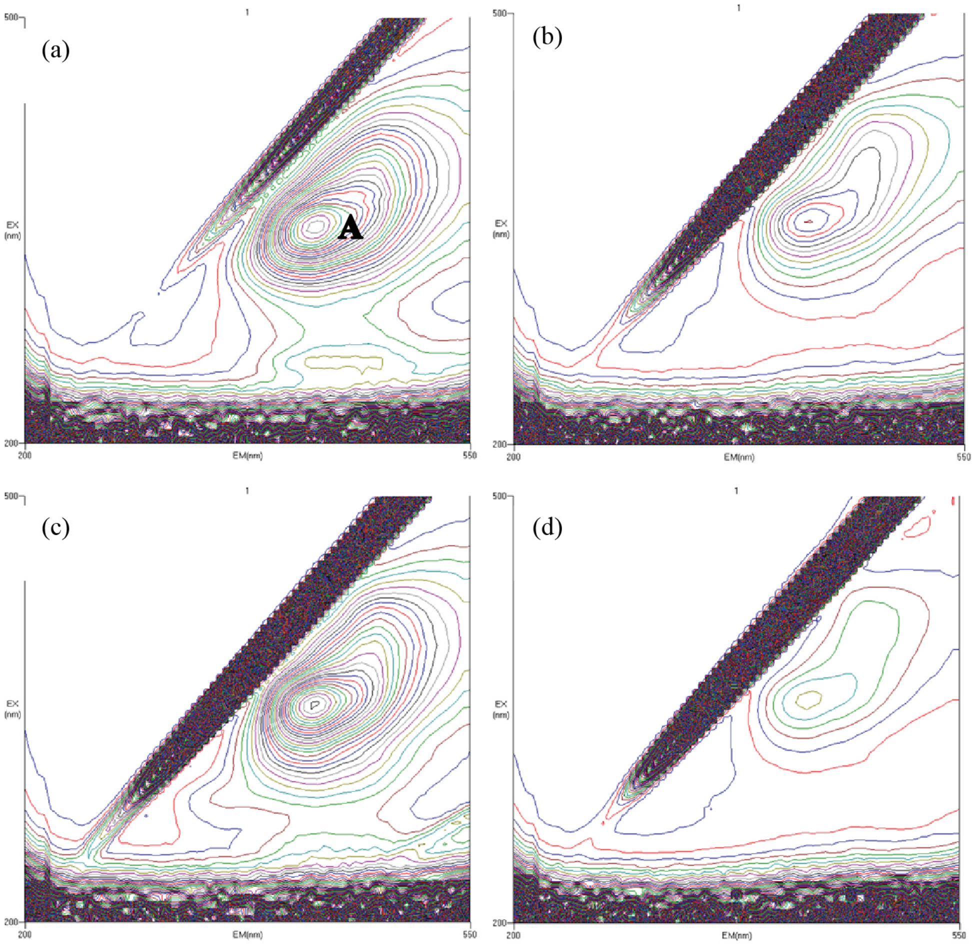

Fig. 3. EEM spectra of (a) MBR-Control SMP, (b) MBR-PP SMP, (c) MBR-Control EPS and (d) MBR-PP EPS.

presented in Fig. 5 MBR-PP had an obvious slower TMP increase rate than MBR-Control. Membrane biofouling is attributed to the two-step biofouling phenomenon, i.e., initial slight TMP increase (first step) followed by a rapid one (second step). Although two MBRs had the similar first step, MBR-PP had an obvious slower TMP increase rate than MBR-Control during second step. Additionally, biofouling resistances (Table 5) were calculated based on the basis of permeation data and resistance-in-series model. R_{total} and R_c both showed the obvious decrease with PAC/PAM flocculation pretreatment. But R_p increased from 5.4 to 6.6 × 10^{12} m^{-1}, which was because of SMP and EPS removal and sludge aggregation, leading to reduce the pore clogging/absorption.

Table 5
Membrane biofouling resistances of MBR-PP and MBR-Control (n = 6)

	Resistance	R_{total}	R_m	R_i	R_p	R_c
MBR-PP	Value (10^{12} m^{-1})	11.9	1.0	0.3	5.4	5.1
	Percentage (%)	100	8	3	46	43
MBR-Control	Value (10^{12} m^{-1})	18.3	1.0	0.3	6.6	10.4
	Percentage (%)	100	5	2	36	57

As above mention, PAC/PAM flocculation pretreatment not only optimized the quality of anaerobically digested swine slurry, but also introduced a certain of Al^{n+} into A/O-MBR, leading to sludge aggregation and further membrane biofouling mitigation. Pendashteh et al. [52] has reported that flocculant of Fe^{3+} and Chitosan could effectively reduce membrane biofouling of MBR due to decreasing SMP, enlarging floc size, increasing RH, etc. As Table 2–4 show sludge aggregation with PAC/PAM pretreatment caused SMP concentration decrease, floc size enlargement and relative hydrophobicity increase, which meant that biofouling mitigation in this study was partly because of sludge aggregation. In addition, Jermann et al. [53] and Xin et al. [54] both reported that multi-valent cations could act as bridges for polysaccharides to enhance gelation, leading to the formation of impermeable gels and further biofouling mitigation. Shen et al. [55] also predicted that flocculation of PAC could remove inorganic and organic components, which partly reduce irreversible fouling. Therefore, the gelation of Al^{n+} caused the polysaccharide aggregation, and transformed into bulk form. Bulk polysaccharide is considered as the negligible role in membrane biofouling process [56]. Therefore, PAC/PAM

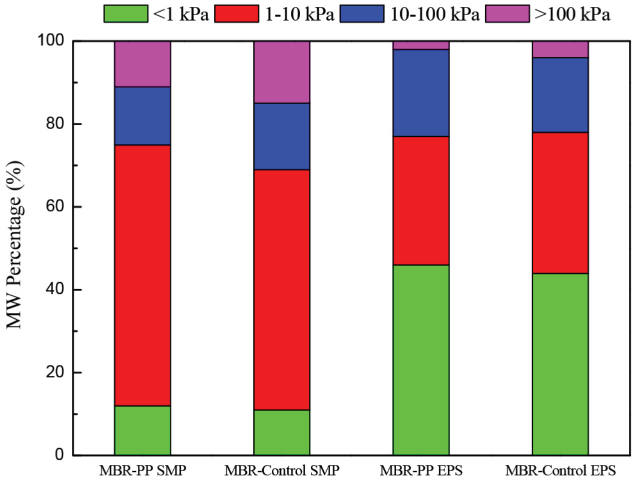

Fig. 4. MW distribution of SMP and EPS in MBRs.

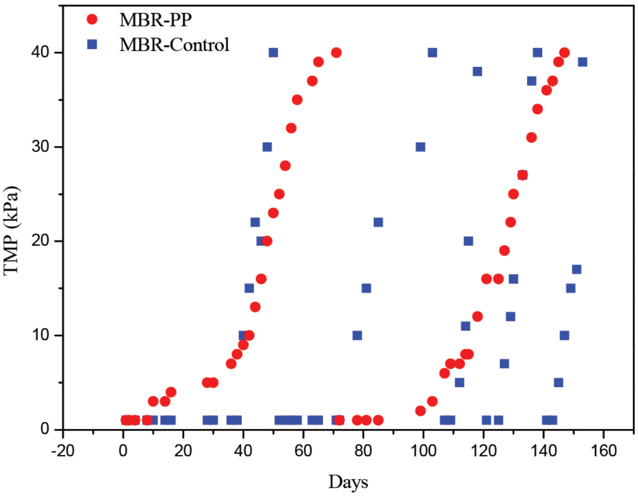

Fig. 5. TMP variations of MBR-PP and MBR-Control.

pretreatment mitigated membrane biofouling, and reduced the total membrane resistance (Table 5). Moreover, Zhang et al. [57] predicted that EPS contributed over 70% drop of the membrane mass transfer coefficient, meaning that EPS directly related to the concentration polarization resistance. Thus, the decrease of concentration polarization resistance was due to EPS decrease. PAC/PAM pretreatment reduced the cake layer resistance of membrane biofouling, and some studies [52,58] reported that large sludge floc slowed down the cake layer formation. It was because the membrane module in this study was transversely set in the middle of the oxic tank, and large sludge flocs easily accumulated on the membrane surface during precipitating.

4. Conclusions

Sludge aggregation and its biofouling mitigation in A/O-MBR treating anaerobically digested swine slurry with PAC/PAM flocculation pretreatment were identified in this study. Flocculation pretreatment with 1600 mg PAC/L wastewater and 50 mg PAM/L wastewater effectively optimized the anaerobically digested swine slurry with obviously COD_{Cr} and TP removal for further biological treatment. Meanwhile, PAC/PAM pretreatment caused approximate 55–74 mg/L Al flowing into A/O-MBR, leading to the obvious sludge aggregation and high density of biomass. PAC/PAM pretreatment also induced SMP decrease and RH increase, and bridged polysaccharides to enhance gelation. EPS decrease with pretreatment further contributed the drop of the membrane mass transfer coefficient. PAC/PAM flocculation pretreatment effectively led to the sludge aggregation and further caused membrane biofouling mitigation.

Acknowledgments

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Supporting Information

Sludge aggregation and biofouling mitigation in A/O-MBR treating anaerobically digested swine slurry with PAC/PAM flocculation pretreatment

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1. Methods and material

1.1. Three-dimensional excitation-emission matrix (EEM) fluorescence spectroscopy analysis

All the samples of EPS and SMP were detected with a luminescence spectrometry (F-4500FL spectrophotometer, Hitachi, Japan). The EEM spectra were analyzed with the scanning emission spectra from 200 nm to 550 nm at 5 nm sampling intervals by varying the excitation wavelengths from 200 nm to 500nm at 5 nm increments (scanning speed = 1200 nm/minutes; the excitation and emission slits = 10 nm). The EEM spectra were plotted as the elliptical shape of contours. The X-axis indicated the emission spectra from 200 nm to 550 nm while the Y-axis expressed the excitation wavelength from 200 nm to 500 nm, and the third dimension, i.e., the contour line, is used to represent the fluorescence intensity at an interval of 5.

1.2. Membrane resistance analysis

The hydraulic resistance was calculated according following equation:

$$R_t = R_m + R_p + R_c + R_i = \frac{\Delta P}{\mu J} \tag{1}$$

where R_m: the constant resistance of the clean membrane; R_p: the resistance due to concentration polarization, R_c: the fouling layer resistance; R_i: the pore blocking resistance; ΔP: TMP; J: permeate flux; μ: viscosity of the permeate water.

The method to measure the resistances of the membrane is as follow:

(1) R_m: the flux and TMP of fresh membrane module are detected in the deionized water before operation.

(2) R_t: the flux and TMP of the membrane module are analyzed according to Eq.(1).

(3) R_p: after the operation, the flux and TMP of the membrane are measured in deionized water according to Eq. (1) to get $R_0 \cdot R_p = R_t – R_0$.

(4) R_i: after removing the cake attached on the membrane surface using physical cleaning, the flux and TMP of the membrane module are measured using deionized water to get R_l according to Eq. (1). $R_i = R_l - R_m$.

(5) $R_c = R_t - R_m - R_i - R_p$.

1.3. Relative hydrophobicity (RH)

25 mL mixed liquor was washed and suspended with 0.1 M phosphate buffer (pH 7.2), and then was ultra sonicated for 2 min. Next, 15 mL n-hexane was added into the suspension and agitated for 10 min. The suspension was transferred to a separatory funnel and waited for 30 min emulsification. The RH was pressed as the ratio of MLSS in aqueous phase after emulsification ($MLSS_e$) to MLSS in the aqueous phase before emulsification ($MLSS_i$):

$$\mathrm{RH}(\%) = 100 - \left(\frac{MLSS_e}{MLSS_i}\right) \times 100$$

Table S1
Characteristics of original anaerobically digested swine slurry[a]

Item	Dissolved COD_{Cr}	Total COD_{Cr}	NH_4^+-N	TN	TP	SS	pH
Value	350–1300	900–3400	400–1100	420–1200	55–95	200–3000	7.4–8.4

[a] pH unit was no unite, and the unit of other items was mg/L.

Table S2
A/O-MBR performance with or without PAC/PAM pretreatment at optimal condition[a]

	Inf. TP	Eff. TP	Inf. NH_4^+-N	Eff. NH_4^+-N	Inf. TN	Eff. TN	Inf. COD_{Cr}	Eff. COD_{Cr}
MBR-PP [b]	17 ± 5	14 ± 3	1050 ± 40	5 ± 4	760 ± 300	680 ± 140	300 ± 40	110 ± 20
MBR-Control [b]	58 ± 12	53 ± 5	1100 ± 60	10 ± 8	760 ± 300	700 ± 160	760 ± 50	680 ± 40
MBR-Raw [c]	78 ± 30	70 ± 25	1100 ± 500	800 ± 500	1400 ± 250	1300 ± 360	2400 ± 800	1800 ± 500

[a] Inf. = Influent; Eff. = Effluent.

[b] TP, NH_4^+-N and COD_{Cr} were measured every 3 days: $n = 50$.

[c] MBR-Raw was operated with similar condition to treat raw swine wastewater for over 60 days in this study. TP, NH_4^+-N and COD_{Cr} were measured every 3 days: n = 20.

[d] Organic loading rate (OLR): MBR-PP = 6000 ± 400 mg COD_{Cr}/m^3 day; MBR-Control = 15200 ± 800 mg COD_{Cr}/m^3 day.

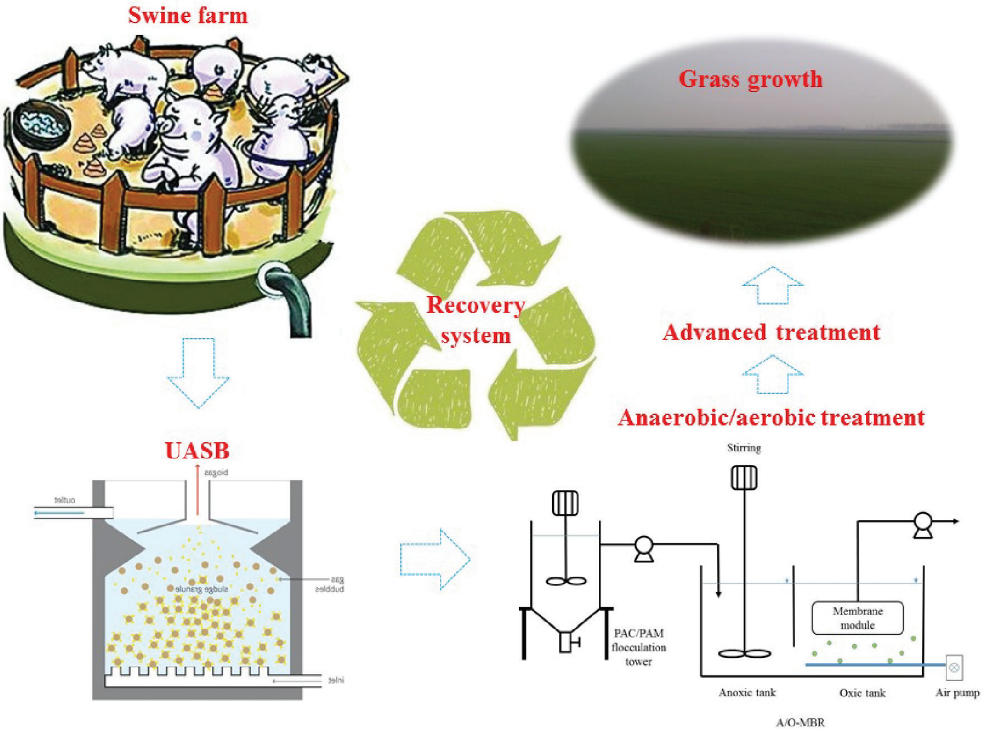

Fig. S1. Recovery of swine wastewater for grass growth. Efficient swine wastewater treatment technology and its engineering demonstration (National Science and Technology Pillar Program: 2013BAD21B03).