

Exerting ultrasound to control the membrane fouling in filtration of anaerobic activated sludge—mechanism and membrane damage

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ABSTRACT

In this study, ultrasound was applied to control membrane fouling development online in an anaerobic membrane bioreactor (AMBR). Experimental results showed that membrane fouling could be controlled effectively by ultrasound although membrane damage may occur under some operational conditions. Based upon the observation on the damaged membrane surface *via* SEM, two mechanisms causing membrane damage by exerting ultrasound are inferred as micro particle collide on the membrane surface and chemical interaction between membrane materials and hydroxyl radicals produced by acoustic cavitations. Not only membrane damage but also membrane fouling control and membrane fouling cleaning were resulted from these mechanisms. Properly selecting ultrasonic intensity and working time, and keeping a certain thickness of cake layer on membrane surface could be effective ways to protect membrane against damage.

Key words | mechanism, membrane damage, membrane fouling control, ultrasound

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INTRODUCTION

Membrane fouling control is a key issue in application of membrane bioreactor (MBR), especially for anaerobic MBR (AMBR). There have been many research papers on membrane fouling mechanisms and control for aerobic MBR. For AMBR, which has great application potential in fields of high strength wastewater treatment and sludge digestion (Daigger *et al.* 2005; Liao *et al.* 2006), there are only limited measures to control its membrane fouling. Hydraulic measure, that is higher crossflow velocity in recirculated AMBR, is most employed with high power consumption (Ross *et al.* 1992; Choo & Lee 1996, 1998). Ultrasound was proved to be able to enhance membrane permeability and mitigate membrane fouling effectively in crossflow filtration of macromolecules (Chai *et al.* 1998; Juang & Lin 2004; Lamminen *et al.* 2004). Ultrasound could also be an effective way to clean membrane fouling in MBR (Chai *et al.* 1999; Li *et al.* 2002; Kobayashi *et al.* 2003; Lim & Bai 2003). However, there have been few studies to

apply ultrasound to online membrane fouling control in filtration of activated sludge.

Our former study on membrane fouling control in an innovated AMBR with ultrasound horns attached to the wall of the membrane module (Figure 1) investigated the proper ultrasound intensity and working time for online membrane fouling control. It showed that the membrane fouling was effectively controlled by exerting ultrasound with power intensity of $0.122 \text{ W}\cdot\text{cm}^{-2}$ and frequency of 28 kHz. However, when the power intensity was $0.203 \text{ W}\cdot\text{cm}^{-2}$, some observations *via* scanning electron microscope (SEM) on the membrane surface showed that the membrane was somehow damaged. The SEM photos also give information about the mechanism of membrane fouling control with ultrasound. This paper is to discuss these experimental phenomena and suggest ways of using ultrasound for both membrane fouling control and protecting the membrane.

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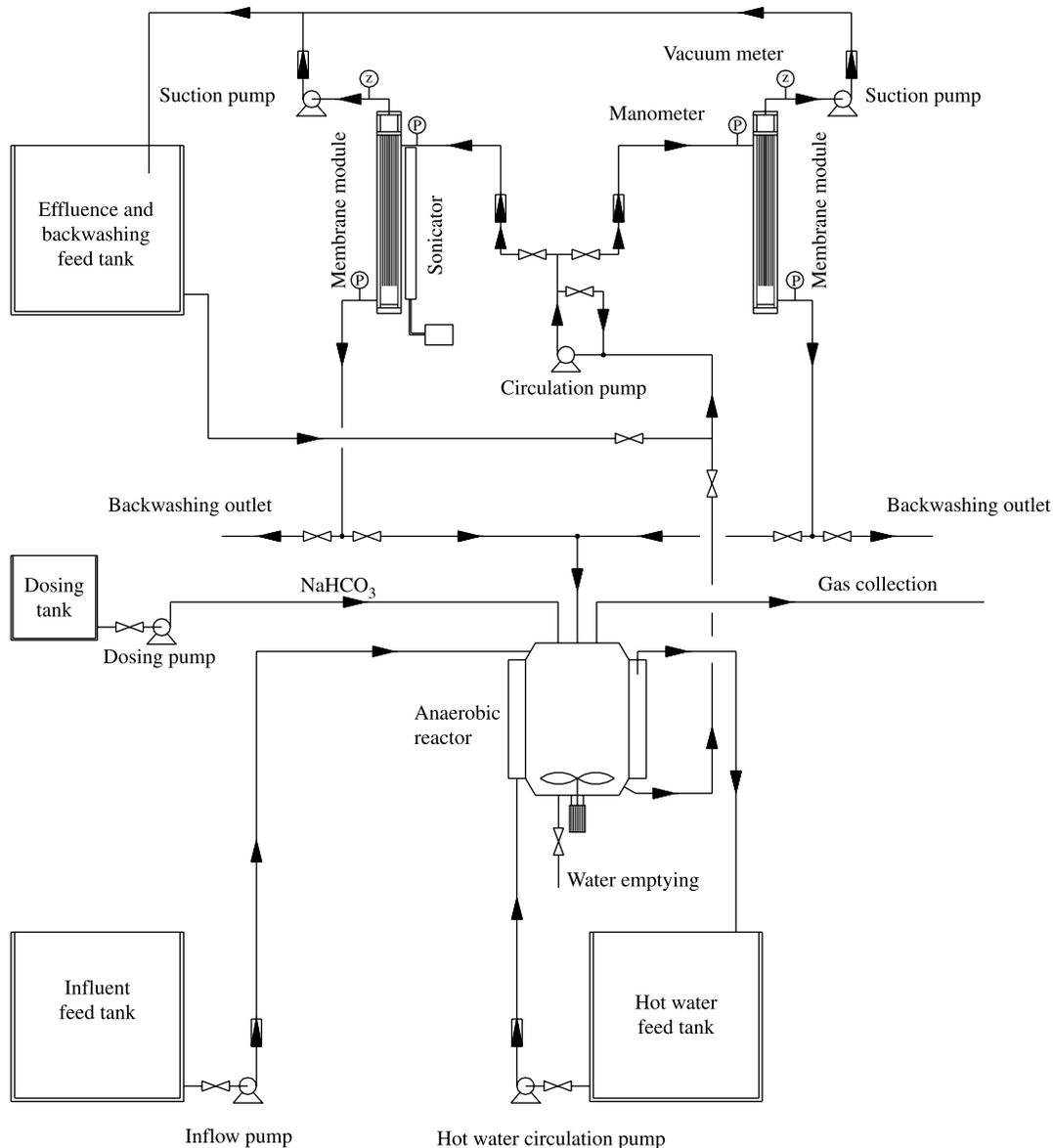


Figure 1 | Experimental setup of anaerobic membrane bioreactor coupled with ultrasonic device.

MATERIAL AND METHODS

The innovated AMBR

Figure 1 shows the schematics of the innovated AMBR coupled with ultrasonic equipment. Two membrane modules were set in the system. One was with six horn ultrasonic transducers, attached to the side face of module, having frequency of 28 kHz and a maximum power output of 300 W. The membrane used in the AMBR

system was hollow fibre polyethylene membrane with pore size of 0.4 μm .

Experimental methods

In this study, the system was operated with a flux of $24\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, crossflow velocity of $0.75\text{m}\cdot\text{s}^{-1}$. Two kinds of ultrasonic power intensity were employed: $0.203\text{W}\cdot\text{cm}^{-2}$ and $0.122\text{W}\cdot\text{cm}^{-2}$ (corresponding to a power output of 150 W and 90 W). Under the condition of ultrasonic power

intensity of $0.203 \text{ W}\cdot\text{cm}^{-2}$, ultrasonic device worked 2 min per 15 min. The influent COD was about 800 mg/L and the sludge concentration was about 3.5 g/L . After 340 hours operation (filtration and ultrasound irradiation), it was found that membrane damage occurred. In order to confirm the size of membrane damage, the particles size distribution in the effluent from the damaged membrane and in the anaerobic suspended mixed liquid were measured. The membrane surface was also imaged *via* optical microscope and scanning electron microscope (SEM, FEI QUANTA200).

Analysis

In this study, membrane fouling was indicated by the total filtration resistance (ΣR , m^{-1}), which is deduced from the Darcy Law.

$$J = \frac{\text{TMP}}{\mu \Sigma R}, \quad \text{and then} \quad \Sigma R = \frac{\text{TMP}}{\mu J}$$

Where, J is permeability flux [$\text{m}^3/\text{m}\cdot\text{s}$]; TMP is trans-membrane pressure [pa]; ΣR is total filtration resistance [m^{-1}]; μ is dynamic viscosity of anaerobic sludge [Pa·s].

The trans-membrane pressure (TMP), flux and dynamic viscosity of anaerobic sludge were monitored in the experiments. Membrane fouling rate ($d\Sigma R/dt$, $\text{m}^{-1}\text{s}^{-1}$) was calculated and used to assess the membrane fouling development.

RESULTS

Membrane fouling control by ultrasound

Figure 2 shows the variations of the total membrane filtration resistance with filtration time. It demonstrates that the membrane fouling was effectively controlled by using ultrasound. After running one week, total filtration resistance of membrane module with ultrasound was less than 25% of that without ultrasound. The filtration was stopped when the SS appeared in the effluent which indicated the membrane was damaged by ultrasound.

Membrane damage

Figure 3 shows the membrane surface observed under optical microscope. It is clear that ultrasound caused the

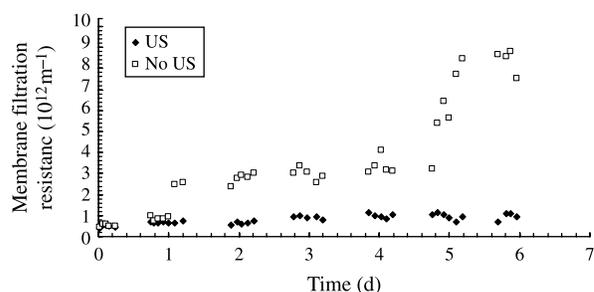


Figure 2 | Filtration resistance variations during filtration course with and without ultrasound (Sludge concentration of $3.5 \text{ g}\cdot\text{L}^{-1}$; Crossflow velocity of $0.75 \text{ m}\cdot\text{s}^{-1}$; Ultrasonic power intensity of $0.203 \text{ W}\cdot\text{cm}^{-2}$; Ultrasound worked 2 min per 15 min).

circle indent on the membrane surface. Figure 4 is the results of the particles size distribution in the effluent from the damaged membrane and in the anaerobic suspended mixed liquid. It could be found that the volume average size of the particles in effluent from the damaged membrane was more than $20 \mu\text{m}$, while that of anaerobic sludge was more than $50 \mu\text{m}$. On the other hand, the pore size of new membrane was only $0.4 \mu\text{m}$ in this study. Furthermore, the volume percentage with the particle size more than $60 \mu\text{m}$ was more than 1% for the effluent of damage membrane. These data indicated that the size of damage hole on the membrane surface was more than $20 \mu\text{m}$, and some reached $60 \mu\text{m}$.

Figure 5 shows the damage on membrane surface observed by SEM. Figure 5a shows the new membrane surface. Figure 5b shows the 500 times magnified images of the damage membrane surface under SEM. It can be seen from Figure 5c to 5f, that two kind of formations occurred on membrane surface, including the circle indents with diameters of $6.25 \sim 40 \mu\text{m}$ (Figure 5c,d) and long and narrow rift with size of about $4 \times 18 \mu\text{m}$ (Figure 5e). Furthermore, a micro particle inlaid on the surface of membrane was found (Figure 5f).

According to images of the membrane under SEM (Figure 5), it could be suggested that in this experiment, following mechanisms were involved in membrane fouling control and damage. One is that micro particle and micro-jets, which obtain high kinetic energy from ultrasonic irradiation, collide with membrane surface. Figure 5f displayed a particle inlaid on the membrane surface, which demonstrated that particle collision took place. The other is chemical oxidation of

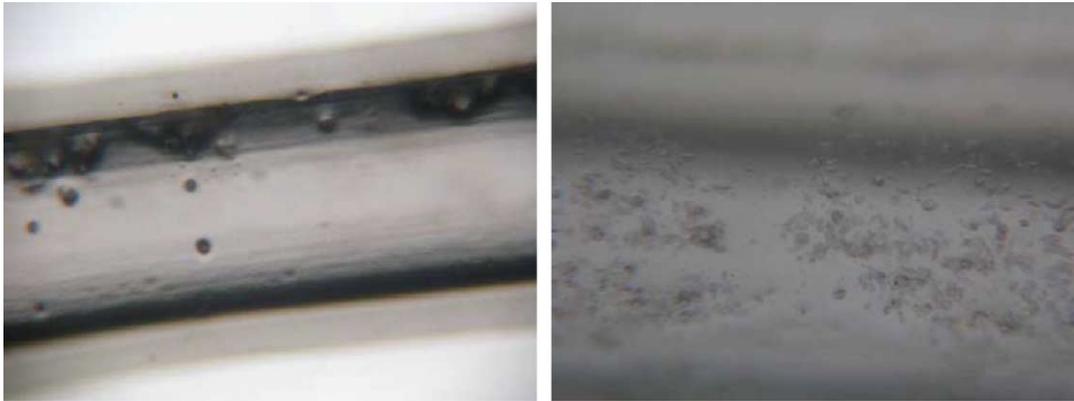


Figure 3 | Damaged membrane surface under optical microscope (left: $\times 100$; right: $\times 400$).

hydroxyl radicals produced by acoustic cavitations caused by ultrasound. This explains the formation of rift with small cracks on its edge on membrane surface showing in [Figure 5e](#), since its formation was totally different with that of hole ([Figure 5c,d](#)), and can not be formed by the particle collision.

The removal of cake layer by ultrasonic cleaning

In this study, application of ultrasound to clean the membrane fouling was also investigated. Under the condition of tap water backwashing crossflow velocity of

$0.75\text{m}\cdot\text{s}^{-1}$ and ultrasonic power intensity of $0.122\text{W}\cdot\text{cm}^{-2}$, ultrasound worked 10 min per 20 min and membrane fouling was cleaned 40 min for removal of cake layer, which formed during one week operation of AMBR.

[Figure 6](#) shows the SEM image of membrane surface before and after ultrasonic online cleaning. The compact cake layer formed on the membrane surface and some microorganism attached on the surface could be observed in [Figure 6a](#), before the ultrasonic online cleaning. However, after online cleaning combined ultrasound and tap water backwashing, cake layer was removed so effectively that some part of cake layer fouling was totally removed and membrane surface could be seen in the image of SEM ([Figure 6b](#)).

DISCUSSION

In this study, the application of ultrasound could control membrane fouling development effectively. However, it also caused membrane damage. By the investigation of ultrasonic cleaning, it was found that cake layer was removed effectively by ultrasound. Theoretically, membrane fouling control, membrane fouling cleaning and membrane damage caused by ultrasound were all resulted from the same mechanism, which could be explained as that ultrasonic irradiations cause physical and/or chemical effects on the mixed liquor when they propagate through it, including heating, compression and rarefaction, turbulence and cavitations ([Masselin *et al.* 2001](#); [Lamminen *et al.* 2004](#)). And then it cause acoustic streaming, micro-jets, micro-streaming, shock wave and chemical interaction with

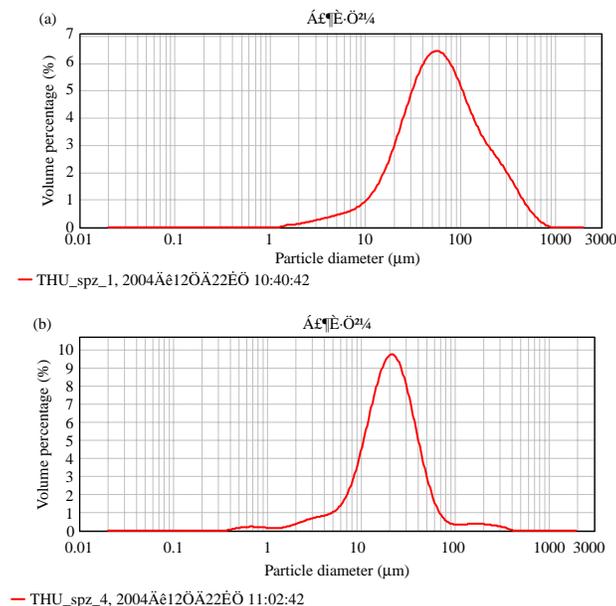


Figure 4 | (a) Particle size distribution of anaerobic suspended sludge (b) Particle size distribution in the effluent of damaged membrane.

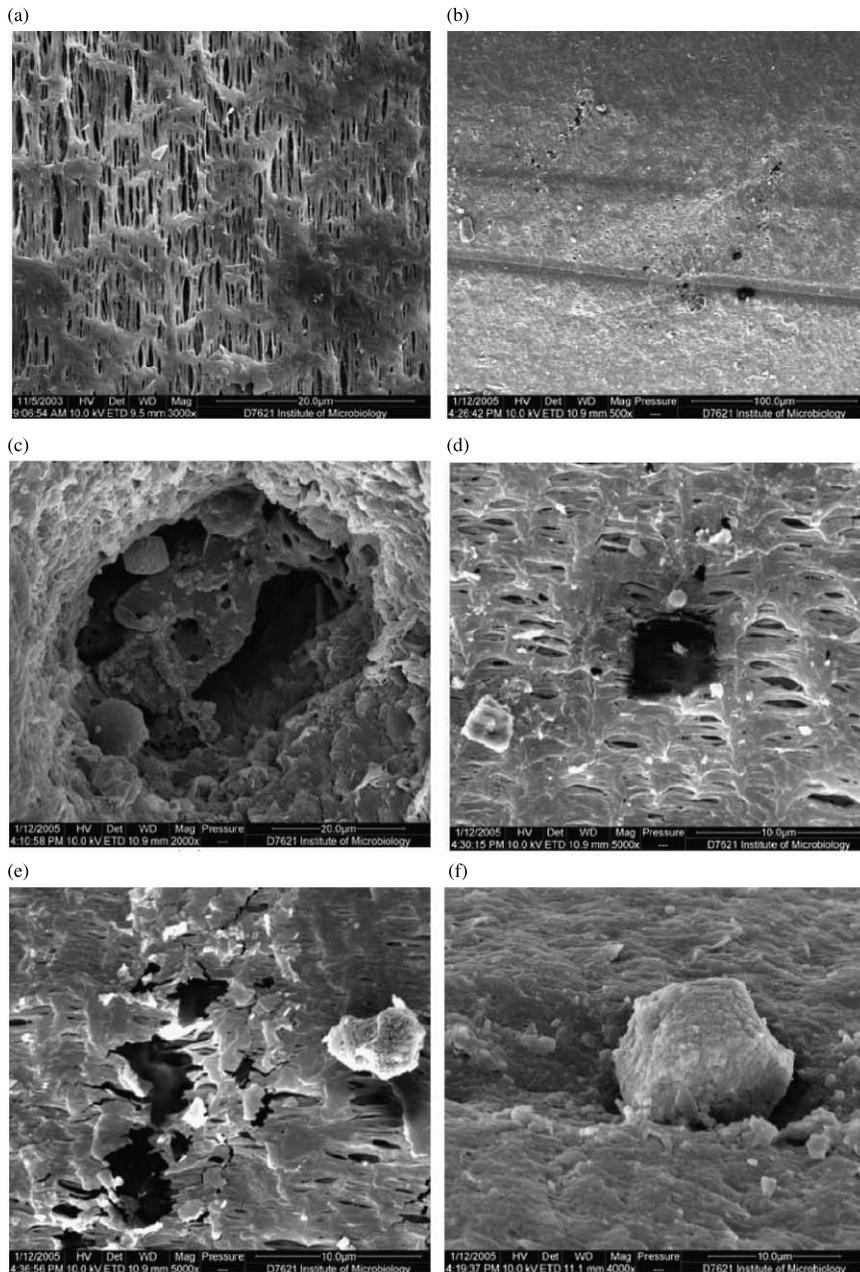


Figure 5 | SEM images of damage membrane surface (a) new membrane ($\times 3,000$); (b) damage membrane surface ($\times 500$); (c) hole ($\times 2,000$); (d) hole ($\times 5,000$); (e) rift ($\times 5,000$); (f) particle inlaid on the membrane surface ($\times 4,000$).

radicals, which have effect on the membrane surface. These effects successively exert force on membrane surface. If fouling layer exists on the membrane surface, these acoustic effects in the liquid will result in the removal of fouling layer; otherwise they will lead to the membrane damage on the naked membrane.

Solutions

Since the membrane damage and membrane fouling control resulted from the same mechanisms, there should be a compromise ultrasound condition to remove membrane fouling and to avoid the membrane damage.

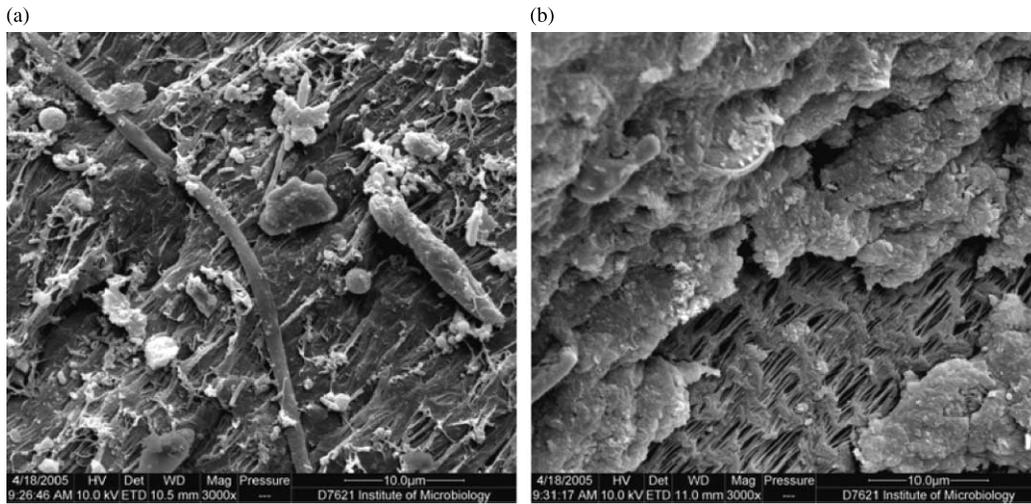


Figure 6 | SEM images of membrane surface before and after ultrasonic cleaning (a) Before ultrasonic cleaning ($\times 3,000$); (b) After ultrasonic cleaning ($\times 3,000$).

In order to avoid membrane damage, ultrasonic fields should be cut down either by decreasing the ultrasonic power intensity or shortening irradiation time. However, in consideration of the membrane fouling, the ultrasonic fields could not be cut down too much. Another way to protect the membrane against damage was to keep a certain thickness of cake layer on the surface of membrane to avoid that the ultrasound has directly effect on the naked membrane surface.

Figure 7 shows a successful demonstration when applying above measures to protect membrane and control membrane fouling. The AMBR was operated under the condition of sludge concentration of $8.0 \text{ g}\cdot\text{L}^{-1}$; influent COD of $1,500 \text{ mg}\cdot\text{L}^{-1}$; crossflow velocity of $0.75 \text{ m}\cdot\text{s}^{-1}$;

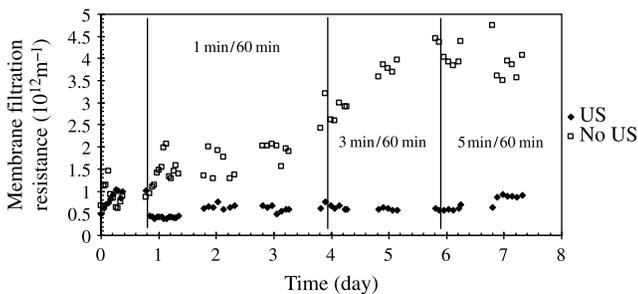


Figure 7 | Filtration resistance variations during filtration course with and without ultrasound working (Sludge concentration: $8.0 \text{ g}\cdot\text{L}^{-1}$; Crossflow velocity of $0.75 \text{ m}\cdot\text{s}^{-1}$; Ultrasonic power intensity of $0.122 \text{ W}\cdot\text{cm}^{-2}$; Ultrasonic work time changed step by step, and 1 min/60 min meant ultrasound worked 1 min per 60 min).

ultrasonic power intensity of $0.122 \text{ W}\cdot\text{cm}^{-2}$. Ultrasonic working time increased step by step from working 1 min per 60 min to working 5 min per 60 min. The results indicated that membrane fouling was controlled successfully. Furthermore, no membrane damage occurred during one week's operations.

CONCLUSION

- (1) Membrane fouling can be effectively online controlled by using ultrasound.
- (2) The major mechanism of membrane fouling control and membrane damage in filtration of anaerobic activated sludge were suggested to be, micro particle collide and micro-jets; and chemical oxidation of hydroxyl radicals produced by acoustic cavitations.
- (3) Combination of ultrasound and tap water backwashing could clean membrane fouling effectively.
- (4) Not only membrane damage but also membrane fouling control and membrane fouling cleaning resulted from the same mechanisms, and properly selecting ultrasonic intensity and working time and keeping a certain thickness of cake layer could be the effective ways to protect membrane against damage.

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