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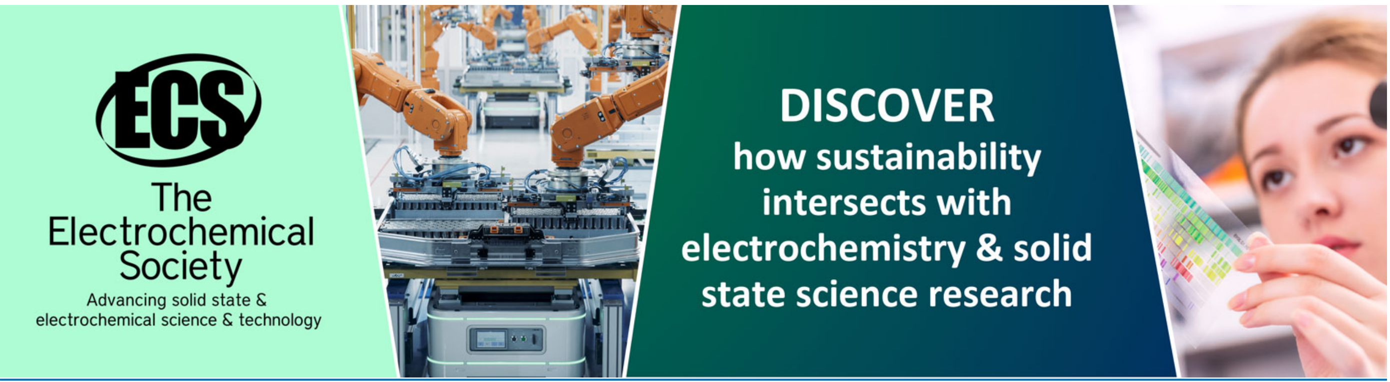

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Anaerobic Membrane Bioreactors for Trace Organic Contaminants with Wastewater Treatment: A Review

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Abstract. Trace organic contaminants (TrOCs) in water environment such as pesticides, personal care products (PPCPs), and industrial materials have the characteristics including low concentration, wide distribution range, stable chemical structure. Anaerobic membrane bioreactor (AnMBR) is a new biological treatment method. The device combines the advantages of membrane filtration technology and biological treatment process. What the performance of AnMBR for TrOCs removal should be noticed. Some research is focusing on this question, and most of research found that AnMBR has shown great promises in the application of the removal of TrOCs in wastewater treatment. This paper briefly overviews recent processes of different types of AnMBR for the removal of different types of TrOCs with the influence of kinds of factors such as temperature, salinity etc. We mainly discussed the construction of AnMBR, the classification of TrOCs, and the removal efficiency of chemical oxygen demand (COD), the removal efficiency of TrOCs, and the biogas production. For the types of AnMBRs, the removal efficiency of COD and TrOCs and biogas production are excellent. Also, the removal efficiency of TrOCs is very related to the types of the contaminants. AnMBR is not only a practical choice for TrOCs removal, but also a good choice for following sustainable development because of the good biogas production. In addition, challenges of the future research are discussed also at the end of this review to give other researchers new perspectives and ideas.

1. Introduction

TrOCs have the characteristics including low concentration, stable chemical structure, long half-life period and obvious bioaccumulation in wastewater. Even with low concentrations, TrOCs in water bodies have toxic and harmful effects on human beings and most aquatic organisms [1-2]. For instance, endocrine disrupting chemical (EDC) is thought to be able to alter normal physiological functions of animal and human by binding natural hormones or ligands or inhibiting natural binding processes, resulting in dysfunction of reproductive system, immune system and nervous system of animal and human gender, and the phenomenon of sex hormone secretion disorder, reproductive capacity decline, gender change and the increase of incidence rate of cancer and other diseases [3].Thus, the removal of TrOCs is important for water pollution control. For the removal and control of TrOCs in water environment, physical and chemical processes have attracted much attention. For example, coagulation, sedimentation, activated carbon adsorption and filtration are widely used in water treatment. The main principle of above methods is to transfer pollutants from water phase to adsorption materials, and further remove pollutants out. Advanced oxidation technology (AOPs) can

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transform TrOCs into small molecules or even complete mineralization by producing hydroxyl radicals with strong oxidation. However, the methods produce side-products. In addition to physical and chemical methods, biodegradation is also used in water pollutants removal, especially used widely in municipal wastewater treatment plants. However, traditional biological treatment methods can only remove easily biodegradable organic pollutants. TrOCs have complex molecular structure and unique biochemical properties are challenge for traditional biological treatment methods [4].

AnMBR is a new technology in wastewater treatment field. It has the advantages of small floor area, saving power consumption, producing biogas and excess sludge at the same time when wastewater treatment [5]. Hydraulic retention time (HRT) and solid retention time (SRT) can be completely separated in AnMBR, which prevents the easy loss of sludge and the disable of methanogens in ordinary anaerobic reactor. This review will briefly introduce the application of AnMBR in wastewater treatment containing TrOCs in published literatures in recent years. The designing of AnMBR, removal efficiencies of TrOCs and COD, as well as the methane yield will be particularly focused.

2. Construction of AnMBR

The structure of AnMBR is mainly combined with an anaerobic bioreactor and a membrane module [6]. There are four types of sub-structure: external cross flow anaerobic membrane bioreactor (ECF-AnMBR), External semi-dead-end filter anaerobic membrane bioreactor (ESD-AnMBR), external anaerobic membrane bioreactor (EMBR) and submerged anaerobic membrane bioreactor (S-AnMBR). ESD-AnMBR and S-AnMBR are the most common types used [7]. There are three forms of membrane module: external cross flow, internal submergence and external submergence [7]. In external cross flow, the membrane is outside the anaerobic reactor, which is easy for cleaning and replacing of the membrane. An external pump is required in this form to recycle the biomass at a relatively high speed. The high speed can wash the membrane surface to reduce membrane pollution and provide high pressure for the liquid passing through the membrane. In the internal submergence form, the membrane is immersed in the anaerobic reactor and the effluent is passed through the membrane by vacuum. The energy consumption of pump transportation can be saved.

3. Applications of AnMBR in TrOCs removal

3.1. Performance of conventional AnMBR for TrOC removal

Huang et al. developed an AnMBR for the treatment of pharmaceutical wastewater containing β-lactams antibiotics (amoxicillin (Amox), ampicillin sodium (Ampt), cefoperazone sodium (Cefo), ceftriaxone sodium (Ceft)) [8]. In this research, a 180 L AnMBR was development with an upflow anaerobic sludge blanket (UASB) reactor and external crossflow UF membrane. The height of the reactor is 1.5 m, and the diameter is 0.4 m. Membrane module was chosen with 1 m^2 surface area and 0.02 μm pore diameter, and work with temperature lower than 40 ℃ and pH in 2-13. At very beginning 16 days, glucose-based wastewater was feeding for starts up, then the wastewater was replaced by pharmaceutical wastewater. When the COD removal efficiency is up to 80%, it means the successful start-up. The whole process was divided into three stages, which HRT is 48.1 h (day 1-60), 35.2 h (day 61-127) and 23.9 h (day 128-253), respectively.

Through the operation of the reactor, the COD content and removal efficiency of each stage has been measured, which indicated that the COD removal efficiency can be improved with higher organic loading rate (OLR). The removal efficiency of the reactor in the whole stage is from 94% to 87.1%. In the first stage, the removal rate of COD is about 91.4%; in the second stage, the removal rate is about 88.14%. Compared with the conventional anaerobic digestion, biogas contains a higher content of methane (56.4-74.6%) in this work. Results showed that the removal efficiency of Cefo was about 74.2% and that of Ampi was about 32.8%. The removal efficiency of Amox and Ceft were 66.9% and 44.8%, respectively. This shows that the type of TrOC has a great influence on the removal efficiency of TrOC. In addition, high effluent quality and low sludge production were obtained in the removal of

TrOCs with AnMBR in this research.

McCurry et al. compared the treatment effect of a pilot-scale anaerobic membrane reactor and a full type aerobic activated sludge system for municipal sewage [9]. The aerobic activated sludge system is from the municipal sewage treatment system. The constructed anaerobic system with a water inflow of 5.5 m^3/day consists of two reactors connected in turn. The first 0.99 m^3 anaerobic fluidized bed reactor contains 139 kg granular activated carbon (GAC); the second 0.77 m^2 anaerobic fluidized membrane bioreactor contains 264 kg GAC. In the aerobic activated sludge system, although some pollutants are not detected in the effluent, the removal rate of carbamazepine (< 20%) and the partial removal rate of sulfamethoxazole (43-86%) are relatively low. For the anaerobic membrane reactor, the removal efficiencies of atenolol (16%), carbamazepine (5%), and trimethoprim (35%) were lower than those of aerobic system, but the removal efficiencies of sulfamethoxazole (88-100%) were higher than those of aerobic system. The erosion of GAC particles on the membrane can control membrane fouling, which is one of the reasons why the performance of anaerobic system is better than that of aerobic system.

3.2. Addition effect for performance of AnMBR

Xiao et al. explored whether addition could affect the TrOC removal efficiency in AnMBR [10]. In the experiment, five types TrOCs was chosen as target, and the effect of the addition of powder activated carbon (PAC) on the treatment efficiency was studied. In this research, an anaerobic membrane bioreactor with an effective capacity of 3.2 L and a 0.11 m^2 microfiltration membrane was established. The temperature was set at 35 ℃. The reactor has been running for 160 days before start-up process. At the end of the start-up process, the wastewater containing trace organic compounds of species medicine was added to the reactor, and the concentration of each antibiotic was 2 μg/L. After the reactor ran stably, PAC was added into the reactor. The SRT was 213 days and the membrane flux were 5 L/ m^2 per h (LMH).

The result for the performance of the AnMBR showed that the removal efficiency of COD is 93.9% before adding activated carbon. After adding activated carbon, the removal efficiency of COD was 93.8%. This shows that the addition of PAC has negligible effect on the removal efficiency of COD. And the output of biogas is about 1.8 L per day with or without activated carbon added, which is also without influence with PAC added. The removal rate of TrOC before and after PAC addition was recorded. Before PAC was added, the remove efficiency of trimethoprim (TMP), sulfamethoxazole (SMX), carbamazepine (CBZ), diclofenac (DCF) and triclosan (TCS) were 94.2%, 92.3%, 9.5%, 22.0% and 47.2%, respectively. In the first five days after adding PAC, the removal efficiency of them was 99% (TMP), 99% (SMX), 90% (CBZ), 88% (DCF) and 90% (TCS), respectively. But as time went on, the removal efficiency of DCF returned to the value before adding PAC. Therefore, it can be concluded that the removal efficiency of the five TrOCs, especially SMX and TCS, can be improved by adding PAC to AnMBR.

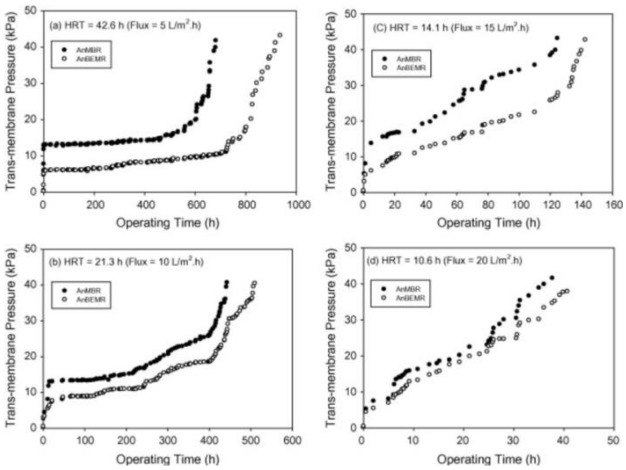

Figure 1. The transmembrane pressure (TMP) changes of both AnMBR and AnBEMR in different HRT with time [11]

Whether adding other substances can improve the performance is also worth considering. Ng et al. constructed a new type of anaerobic bio-entrapped membrane reactor (AnBEMR) and compared the performance with AnMBR [11]. Two laboratory-scale anaerobic membrane reactors were established. Anaerobic embedded ball is used in one reactor, which can be called AnBEMR. About 70 day later, AnMBR and AnBEMR completed the acclimation period. The removal efficiency of TCOD of two AnMBR was less than 45%. At the end of the period, the methane production was 73.0-133.1 mL CH_4/g COD. The methane content in the biogas produced by AnMBR and AnBEMR is 58.8-72.9% and 61.1-72.1% respectively. Therefore, the methane production of AnBEMR (about 15%) is higher than that of AnMBR. Both AnMBR and AnBEMR have poor removal efficiencies of organics, which may be due to the high HRT, the complex organics and the high salinity in pharmaceutical wastewater. The effect of two different reactors on TMP was also investigated in this research. Figure 1 shows the change rule of TMP in two reactors under different HRT. When HRT is large, and AnBEMR showed a stronger ability to resist membrane fouling. However, when HRT was very small, the membrane fouling problem of both reactors occurred very early. This work shows that HRT and membrane pollution had a reverse relationship, and the addition of bio-ball can affect the membrane pollution.

Whether the difference on the membrane will affect the removal efficiency is also a question worthy of consideration. Therefore, Wei et al. used anaerobic membrane reactor to treat 15 TrOCs, in which ultrafiltration and nanofiltration were respective used and the treatment efficiencies were compared [12]. A 2 L laboratory-scale anaerobic membrane reactor was established, and the condition set with 35 °C, pH=7, HRT=12 h, COD=400 mg/L and flux of 6 L/m^2/h. The whole reactor has been maintained for 4 months with two phases divided before the whole experiment. The concentration of each TrOCs in phase 1 (1-30 days) is 10-20 μg/L, and UF membrane module with 30 nm pore size and 310 cm^2 filtration area is used in the whole reactor. Phase 2 (31-70 d) is added into a nanofiltration (NF) system to treat 4 L of wastewater every day, of which 2 L is discharged, and the other 2 L is returned to the reactor. OLR and OMPs maintained as 0.8 g COD/L/d and 20-40 μg/L/d respectively in Phase 1. On the last ten days of phase 2, PAC was added to the reactor. In phase 1, the removal rate of COD is 97%; in phase 2, the removal rate of COD is 97% to 92%, but in the whole AnMBR-NF system, the removal rate of COD is 99%. After adding PAC, the initial COD concentration decreased, but then increased immediately, indicating that PAC has only limited adsorption capacity. Biogas's output is very stable, and the ratio of metal is very stable in the range of 70% - 80%. It can be concluded that NF has no effect on biogas production.

There is no intrinsic correlation between the biodegradability and application of organic matter pollutants (OMPs). In bioreactor, the characteristics of compounds are the key factors affecting their biodegradability and removal rate, among which molecular hydrophobicity plays an important role and the degradation rate is significantly higher than that of hydrophilic molecules. After adding NF, the total removal rate of all compounds in AnMBR-NF is better than that of AnMBR alone, especially the refractory compounds. It shows that NF has direct rejection to OMPs, thus the reactor has more time to degrade the organic pollutants which are difficult to degrade. When PAC was added, the removal rate of COD and OMPs increased, but after a period of time, the removal rate of COD and OMP returned to the state before PAC added, indicating the inadequate adsorption capacity of PAC for bulk organics.

High sulphite concentration in water can inhibit the activity of anaerobic microbial communities [13]. Therefore, Kaya et al. explored the effect of pre-ozonation treatment on removal efficiency of TrOCs with AnMBR [14]. A 160 cm diameter, 320 cm height reactor was chosen and microfiltration membrane with an area of 66 cm^2 and hole size of 0.05 μm was used. The ozonation reaction was carried out in a 2 L stainless steel vessel in one hour under the condition of 35 °C and pH of 7. The ozone treatment was carried out in lab scale with 2 g/h ozone produced. The whole experiment is divided into three stages. A flat membrane and no pre-ozonation treatment were performed in first stage with 390 days, and COD concentration increased from 2500 to 15000 mg/L. The second stage was 167 days and pre-ozonation treatment was added to the whole experiment. The third stage is 168 days, which changed flat membrane to hollow fibre membrane, and ozone pre-treatment also continues to be used.

The experimental results show that the COD removal rate decreases from 84% to less than 50% in first stage, which indicates that the accumulation of sulphite leads to a sharp decline of COD removal efficiency. In the second and third stages, with the addition of pre-ozonation treatment, COD removal efficiency finally recovered to 90%. This shows that pre-ozonation treatment can effectively remove the sulphite content in the reactor and improve the removal efficiency of COD. In the first stage, the removal rate of etodolac was 15% to 49% and finally decreased to 5.4%. However, in the second and third stages of ozone pre-treatment, the removal efficiency of etodolac reached 90.1% and 99%, respectively. Therefore, the ozone pre-treatment process can effectively improve the biodegradability and removal efficiency of etodolac and the removal rate of COD.

3.3. Molecule and environmental effect

Wijekoon et al. reported the relationship between properties of 27 TrOCs and removal efficiency of TrOCs with AnMBR [15]. In this research, a 30 L stainless steel reactor was established, and an external ceramic membrane was set up. One conductive level controller is used to maintain a stable working capacity of 20 L in the reactor. The pore diameter and effective plane size of the ceramic membrane are 1 μm and 0.09 m^2, respectively. The pH of the bioreactor was kept at 7 and the temperature stayed at 35 ℃. HRT, membrane flux and OLR were 4 days, 1.8 L/ m^2 h and 1.3 g COD/L per day, respectively. SRT is about 180 days. The selection of TrOCs is mainly based on the probability of their existence in wastewater, the hydrophobicity and molecular structures, including personal care products, pharmaceuticals, pesticides and industrial chemicals. The concentration of each TrOCs is 5 μg/L. To measure the concentration of TrOCs in water phase, high performance liquid chromatography (HPLC) was used.

When TrOCs was added, the removal efficiency of total nitrogen and COD and the production rate of biogas had some changes, but after two weeks, they stayed stable. In treatment process, the removal efficiency of COD is 84% and that of TN is less than 20%. Therefore, the removal efficiency of TN in this reactor can be ignored. Methane production is 0.2 L CH_4/gCOD. Methane accounts for 61% of the total biogas. Biogas is basically stable at 5.4 L/d. The results which are figure 2 show that the removal rate of TrOCs is closely related to hydrophobicity. When the log D is greater than or equal to 3.2, which means the molecule is high hydrophobicity, the removal rate of TrOCs is higher than 70%. When the log D is less than 3.2, the removal rate of TrOCs changes greatly. The reason may be related to the molecular properties and the functions of TrOCs. Researchers divided TrOCs into three groups for comparison. Group A contained hydrophilic compounds (log D ph7 < 3.2) and at least one electro donor functional group (EDG), but no electron withdrawing functional group (EWG); group B contained hydrophilic compounds (log D ph7 < 3.2) and at least one EWG functional group; group C contained hydrophobic compounds (log D ph7 < 3.2). Results in Figure 2 showed that hydrophobic pollutants could be well removed, and the removal efficiencies of hydrophilic TrOCs are poor and could accumulate in sludge phase. Otherwise, it can also help researchers to separate and identify specific TrOCs characteristics.

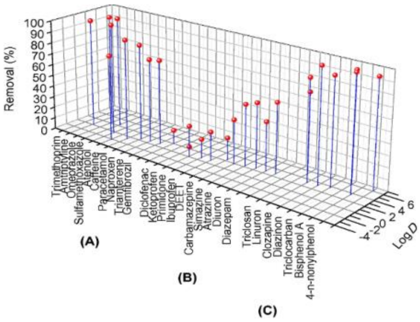

Figure 2. Relationship between Log D and removal efficiency of TrOCs [15]

Hu et al. created a novel AnMBR to evaluate the performance [16]. In this research, the removal efficiency of THF by AnMBR, the effect of volume loading rate (VLR) on the removal rate of the fermentation products and microbial structure were analysed. An internal circulation (IC) was consisted with a diameter of 1.5 m and a height of 2.5 m and a hollow fibre membrane module with an area of 20 m^2. The reactor was operated at 40 ℃ and pH 2-13. The membrane flux was 20 L/(m^2 h) The HRT was 48 h and the COD concentration in the influent water was 1000 mg/L. When the reactor was stable, the COD concentration was continuously added to 20000 mg/L. The temperature is 35 ℃ and TMP is controlled at 60-80 kpa. The whole experiment lasted 249 days. DGGE and T-RFLP were used to analyse the microbial structure.

With the COD concentration increased in each phase, the COD removal rate slightly decreases, and recover to more than 95% immediately. In phase five, the COD removal rate decreased to 80% with the high influent COD concentration of 20000 mg/L. This result indicates that the COD concentration has exceeded the maximum metabolic acceptance range of microbial community in the reactor. The removal rate of THF remained stable at 99%, and not influenced with change of COD concentration. The biogas yield of Phase1 was 94 L/d. This is because the microorganisms in the reactor are still in the cultivation stage. From phase 2 to phase 4, the biogas production remained about 130.7 L/d, but decreased to 68.3 L/d in phase 5. This indicates that high concentration of COD will inhibit the metabolism of anaerobic microorganisms in the reactor. Four microorganisms were observed in this reactor. With the increase of the concentration of methoxazole, the concentration of methanobacteriales is close to 0. With the continuous increase of concentration, the relative abundances of methanosaeaceae decreased at phase 5, causing the methane production also decreased at phase 5.

Cross flow velocity (CFV) is the transverse velocity on membrane surface, which is an important parameter of membrane bioreactor. Whether CFV affect the removal efficiency of TrOCs in AnMBR is unknown. Therefore, Hu et al. set different CFV, to explore the influence on operational efficiency [17]. Tetrahydrofuran (THF) is a common TrOC in municipal wastewater, so this experiment selected THF as the represent of TrOCs. A pilot bioreactor is divided into two parts: an internal circulation which has a height of 2.5 m, a diameter of 1.5 m and a volume of 4.4 m^3 and a membrane module which has an area of 20 m^2 and a length of 1850 mm, and work with temperature lower than 40 ℃ and pH in 2-13. The membrane flux is 20 L/(m^2 h) and HRT is 48 h. The source of COD in the influent water is glucose. The specific operations of THF are THF 200-4500 mg/L, influent cod 1000-25000 mg/L. The whole experimental process is divided into four running times, in which the CFV of each time is 0.7 ±0.1, 1.5 ±0.1, 2.1 ±0.1, 1.5 ±0.1, respectively.

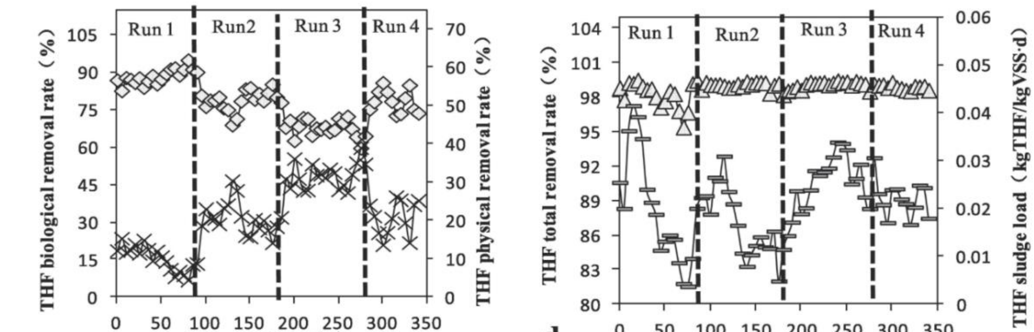

Figure 3. THF removal efficiency with different running time [17]

After 341 days of operation, the COD removal efficiency of the four runs were calculated as 97%, 96.3%, 96.3% and 96.5% respectively. Therefore, with the increase of CFV, from 0.7 to 2.1, the removal efficiency of COD did not increase or decrease significantly. Therefore, the value of CFV does not affect the removal efficiency of COD. The Figure 3 shows the removal efficiencies of THF in different runs. As shown in Figure 3(a), with the increase of CFV, the biological removal efficiency of

THF is decreasing, but the physical removal efficiency of THF is increasing. Therefore, CFV affects the removal of THF. However, according to Figure 3(b), the total removal efficiency of THF remains unchanged in four runs. Therefore, CFV does not affect the removal efficiency of THF. In a word, CFV does not affect the removal performance of COD and THF, but CFV will affect the removal mode of THF. With the increase of CFV, the physical removal efficiency of THF will increase, but the chemical removal efficiency will decrease.

A feasible technology for enhancing removal of TrOC by AnMBR is to pre-concentrate the COD in the municipal sewage to a suitable range for wastewater treatment. This technology can be achieved by using forward osmosis or other high rejection membrane processes to extract clean water directly from municipal sewage and produce concentrated sewage solution. However, the pre-concentration process before AnMBR leads to the increase of salinity in the concentrated municipal wastewater. And the effect of high salinity on the performance of anaerobic membrane bioreactor is few to known. Song et al. explored the effect of high inorganic salt concentration on the basic biological performance of AnMBR and the removal efficiency of 33 types of TrOCs [18]. They developed a 30 L bioreactor with an external ceramic microfiltration membrane. The pore size and the effective area of the membrane are 0.1 μm and 0.09 m^2, respectively. The reactor temperature was maintained at 35 ± 1 ℃. After COD removal rate was more than 96%, start-up process was completed, and TrOCs were added with concentration of 2 μg/L. The sodium chloride concentration was increased from 0 to 15 g/L, following a daily increase of 1 g/L. The influent salt concentration was maintained at 5, 10 and 15 g/L NaCl for two weeks, respectively. HRT was 5 days, and pH stayed at 7. The flux was 1.8 L/(m^2 ·h)

In the whole treatment process, the removal efficiency of TOC was about 98%. Therefore, it can be concluded that the influence on TrOC removal is negligible with the increase of the salinity. However, with the salinity of bioreactor changed from 0 to 10 g/L, the removal efficiency of COD decreased from 98% to 80% was observed. In addition, only a small decrease of biogas was observed with the salinity increased to above 10 g/L. The removal efficiency of hydrophobic TrOCs by AnMBR was basically higher than 80% (log D > 3.2, pH 7). The change of salinity has little effect on the removal efficiency of hydrophobic TrOCs. However, the removal rate of hydrophilic TrOCs (log D < 3.2 at pH 7) changed significantly. Therefore, high salinity in AnMBR can reduced the removal rate of most hydrophilic compounds but had little effect on the removal of hydrophobic TrOCs.

4. Conclusion

This paper mainly discusses and reviews the removal efficiency of TrOCs by AnMBR. Recent studies have shown that AnMBR have high COD removal efficiency (basically more than 85%) and high biogas production, but the removal efficiency of TrOCs is very different. One of the reasons is that the physical and chemical properties of TrOCs have an important impact on the removal efficiency. The removal efficiency of the hydrophobic molecules (log D > 3.2) can be as high as 70%. The composition of the influent water, such as COD concentration, will affect the removal efficiency of TrOCs. The difference of environment in the reactor (such as salinity) also affects the removal efficiency of TrOCs. Adding GAC, PAC, bio-ball or ozonation pretreatment before wastewater enters the reactor can significantly improve the removal efficiency of TrOCs. The different use of membrane also affects the working effect of the whole reactor. Nanofiltration membrane module has greatly improved the working efficiency of the whole reactor. Therefore, adding nanofiltration membrane to improve the removal efficiency of TrOCs is a recommend choice. All in all, it can be concluded that AnMBR can be a very promising technology in TrOCs removal. Future research should focus on the control of membrane fouling, the reduction of cost, and the combination of AnMBR with other technologies. This review will provide useful guidance for researchers working in the application of AnMBR in the removal of TrOCs.

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