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厌氧膜生物反应器处理含盐废水运行效能及膜污染特性

闫欢汐¹, 许振钰¹, 金春姬^{1,2*}, 邵梦雨¹, 郭亮^{1,2}, 赵阳国^{1,2}

(1.中国海洋大学环境科学与工程学院,青岛 266100; 2.中国海洋大学海洋环境与生态教育部重点实验室,青岛 266100) 摘要:采用厌氧膜生物反应器(anaerobic membrane bioreactor, AnMBR)处理模拟含盐有机废水,研究盐度变化对反应器运行效能及膜污染特性的影响.结果表明,当进水盐度逐渐增加但低于9.1 g·L⁻¹时,反应器运行稳定,出水效果良好;当盐度增加至10 g·L⁻¹,反应器 COD 去除率、产气量及甲烷含量都明显下降;污泥浓度、污泥体积指数(SVI)、溶解性微生物产物(SMP)和胞外聚合物(EPS)均随盐度增加而先升后降,污泥絮体紧密,沉降性良好.中空纤维膜组件在118 d内运行了3个周期,随着盐度增加膜运行周期由31 d 延长为48 d,膜污染有所减缓.用 SEM-EDX 分析发现膜面污染物中有类似结晶状物质,Na、Mg、Al、Si、Cl、K、Ca和Fe为主要无机元素.三维荧光光谱(EEM)分析表明,蛋白质与腐殖酸是膜面有机污染物的主要成分.

关键词: 厌氧膜生物反应器(AnMBR); 盐度; 膜污染; 污泥性质; 污水处理

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Performance and Membrane Fouling Properties in an Anaerobic Membrane Bioreactor for Salty Wastewater

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Abstract: In the research, an anaerobic membrane bioreactor (AnMBR) was used to treat simulated salty organic wastewater, and the effect of salinity on reactor performance and membrane fouling properties was investigated. The results indicated that when the influent salinity increased gradually but was lower than $9.1~\rm g\cdot L^{-1}$, the reactor ran stably and the effluent performance was good. When the salinity increased to $10~\rm g\cdot L^{-1}$, the COD removal-efficiency, gas production, and methane content decreased significantly; meanwhile, the sludge concentration, sludge volume index (SVI), soluble microbial products (SMP), and extracellular polymeric substance (EPS) levels became elevated at first and then declined with the rising salinity. The system developed compact flocs and a high settling ability. The hollow fiber membrane module was run for three cycles in 118 d. The membrane operating cycle was extended from 31 d to 48 d with the increasing salinity, which favored the control of membrane fouling. SEM-EDX analysis results revealed that there were similar crystalline substances in the film membrane foulants, and Na, Mg, Al, Si, Cl, K, Ca, and Fe were the main inorganic elements. Excitation emission matrix (EEM) analysis results demonstrated that proteins and humic acids were the main components of the organic membrane foulants.

Key words: anaerobic membrane bioreactor(AnMBR); salinity; membrane fouling; sludge properties; wastewater treatment

近年来,石油、印染、化工、皮革等行业快速发展,随之而来的含盐废水量急剧增长[1].这类废水通常采用传统生物法进行处理,但由于其含有的高浓度无机盐物质会造成细胞溶解、生物质呼吸速率降低、沉降性变差,使得传统生物法处理效率低下^[2].针对此类废水处理难度大、流程复杂、成本过高等问题,寻找一种经济高效、简单实用的处理技术才能满足日趋严格的行业污水排放标准.

厌氧膜生物反应器(anaerobic membrane bioreactor, AnMBR)是厌氧生物处理与膜分离技术相结合的污水处理技术,该技术通过膜过滤截留作用,增加污泥浓度和污泥停留时间,有利于积累对盐度耐受的微生物^[3]. Yurtsever等^[4]采用 AnMBR处理含有不同质量浓度(0~1000 mg·L⁻¹)NaCl 的纺织合成废水,发现尽管生物质浓度明显降低,但COD 去除率仍高达90%以上; Jeison等^[5]分别用传

统和装有膜的上流式厌氧污泥床反应器处理含盐废水,在 Na 质量浓度由 24 g·L⁻¹降至 16 g·L⁻¹时,传统污泥床反应器内挥发性脂肪酸(VFA)浓度骤增,反应器内严重酸化,装有膜的反应器则运行稳定.这说明 AnMBR 在处理高盐高有机物废水时,不仅稳定性良好,而且表现出较高的耐冲击负荷能力,拥有广阔的发展前景^[6].

厌氧膜生物反应器在实际工程中的应用受到局限的主要原因是膜污染^[7]. Huang 等^[8]利用浸没式AnMBR 处理生活污水,发现膜污染在 SRT 为 60d时得到最佳控制,延长或缩短 SRT 都会导致更多的颗粒物积累在膜表面,增加膜污染. 原晓玉等^[9]在

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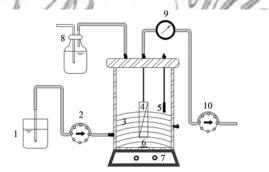
25℃和 35℃下分别运行 AnMBR,观察到温度为 35℃系统膜污染速率更低,膜运行周期更长. Huang 等^[10]的研究表明较短的 HRT 会促进生物量和可溶性微生物产物(soluble microbial products, SMP)中蛋白质类物质的增长从而加速膜污染. 目前对 AnMBR 在处理含盐废水条件下引起的膜污染特性研究还较少见.

本实验采用 AnMBR 处理模拟含盐有机废水, 重点考察提升盐度对反应器运行效能、产气能力、 污泥性质、膜污染特性的影响,以期为厌氧膜生物 反应器在实际处理含盐废水工程中的应用提供理论 依据.

1 材料与方法

1.1 实验装置

本实验装置如图 1 所示, AnMBR 反应器主体是有机玻璃材料构成的圆柱体, 高 25 cm, 直径 22 cm, 有效容积为 8 L. AnMBR 设置温控装置,通过电热丝将温度控制在(35 \pm 0.5) $^{\circ}$ C. 由聚偏氟乙烯(polyvinylidene fluoride, PVDF)制成的中空纤维膜组件,膜表面积 0.235 m², 平均孔径 0.22 μ m, 以悬挂方式置于污泥混合悬浮液中.



配水箱; 2. 进水泵; 3. 加热丝; 4. 中空纤维膜组件;
 温控探头; 6. 搅拌子; 7. 磁力搅拌器; 8. 气体收集装置;
 9. 压力表; 10. 出水泵

图 1 AnMBR 反应器装置及示意

Fig. 1 Schematic diagram of the AnMBR

本实验进水通过蠕动泵流入反应器,利用磁力搅拌使之与污泥悬浮液混合均匀,水力停留时间设置为 6 d. 反应器外部设有气体收集装置,采用排水法收集厌氧反应产生的沼气. 膜组件与出水泵间设置真空表监测跨膜压差(transmembrane pressure,TMP),当 TMP 超过 35 kPa 时,对膜组件进行物理化学清洗.

1.2 污泥与进水水质

本实验所用接种污泥取自青岛市麦岛污水处理厂,接种污泥的混合液悬浮固体浓度(mixed liquid suspended solids, MLSS)为31.9 g·L⁻¹,接种体积

1 L. 反应器进水采用配制的模拟含盐有机废水, 其组成为: $C_6H_6O_6$ 、 CH_3COONa 、 NH_4Cl 和 KH_2PO_4 ,按 COD: $N: P \approx 55: 5: 1$ 比例配制成模拟含盐有机废水. 盐度采用投加海水晶控制,由于海水晶含有 Ca^{2+} 、 Fe^{2+} 、 Mg^{2+} 、 Mn^{2+} 、 Zn^{2+} 和 Co^{2+} 等离子,因此进水不额外添加微量元素.

1.3 出水水质与污泥性质分析方法

COD 采用重铬酸钾法测定; pH 采用雷兹 PHS-3DpH 计测定; VFA 和气体采用气相色谱法测定; 混合液悬浮固体浓度(MLSS)和混合液挥发性悬浮固体(mixed liquor volatile suspended solids, MLVSS)采用重量法测定; 蛋白质采用 Folin-酚试剂法^[11]; 多糖采用苯酚-硫酸法测定^[12].

1.4 膜污染特性分析方法

出水时启动蠕动泵, 待真空压力表读数稳定后, 将该读数记为当天跨膜压差(TMP)值, 同时收集一定体积水量并记录耗时, 即可计算当天膜通量和膜总阻力. 膜总阻力根据 Darcy 公式[式(1)]^[13]计算:

$$R = \frac{\Delta P}{\mu J} \tag{1}$$

式中, R 为膜总阻力, m^{-1} ; ΔP 为跨膜压差(TMP), $Pa; \mu$ 为渗滤液动力黏度系数, $Pa \cdot s; J$ 为膜通量, $L \cdot (m^2 \cdot h)^{-1}$.

膜面污染物特征及其元素组成采用扫描电子显微镜(SEM)和能量散射 X 射线能谱仪(EDX)分析,膜面污染物有机成分采用三维荧光光谱(EEM)分析.

2 结果与讨论

2.1 AnMBR 运行特性

2.1.1 盐度对 COD 去除效果的影响

AnMBR 运行分为两阶段,包括 54 d 启动阶段和 64 d 盐度提升阶段.启动阶段进水 COD 从 900 mg·L⁻¹逐步上升并稳定在2 100 mg·L⁻¹.如图 2 所示,反应器对 COD 的去除率在初期因为水力停留时间(HRT)过短而出现剧烈波动,调整 HRT 后,COD 去除率提高至 97%以上.盐度提升阶段,在一定盐度范围内 COD 去除率随进水含盐量增加略有上升,说明适当的盐度通过调节微生物渗透压和酶活性,提高了微生物利用有机物的能力[14];COD 去除率在盐度增加到 9.1 g·L⁻¹时急速下降,此后虽然延长了每个盐度适应期,但出水 COD 浓度仍在增加,COD 去除率最终在盐度为 10 g·L⁻¹时降至80%以下,这可能是因为较高的盐浓度使微生物胞外渗透压过高,导致细胞脱水产生质壁分离、细胞

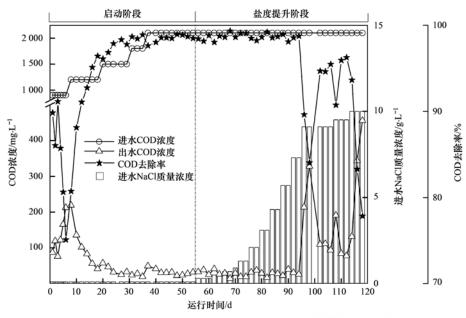


图 2 AnMBR 对 COD 去除效果

Fig. 2 COD removal in the AnMBR

质溶解并最终裂解死亡[15].

2.1.2 盐度对 pH、VFA 的影响

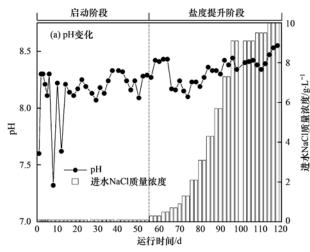
在厌氧处理过程中,产甲烷菌的最适 pH 范围在 6.8~7.2之间,当进水 pH 在上述范围内时,分析图 3(a)发现反应器运行过程中出水 pH 除在启动初期出现波动外,平均值在 8.0~8.5之间,表明反应器内无酸化现象发生.

挥发性脂肪酸(VFA)浓度是判断厌氧反应器性能特别是产酸菌和产甲烷菌活性的重要指标^[16].如图 3(b)所示, VFA 含量随有机负荷增加而降低并稳定在 150 mg·L⁻¹左右,这与 Jeison等^[17]提高有机负荷产生更多量 VFA 的研究结果不同,分析原因可能是其反应器酸化随有机负荷增加升高达65%,而本实验中 AnMBR 运行良好未发生酸化现象,大量的有机酸被分解转化成为甲烷、二氧化碳

等物质. 盐度提升阶段,短期盐度增加使产甲烷菌利用 VFA 的能力下降, VFA 最高增加到 342 mg·L⁻¹; 当盐度在 0.9~9.1 g·L⁻¹范围内增加时, VFA 总量下降且乙酸在 VFA 总量中的占比明显提高,而在所有 VFA 中,乙酸和丁酸最有利于甲烷形成,其中乙酸的贡献超过 70% [16],这说明盐度在这一范围内变化时能促进产甲烷菌将挥发性有机酸转化为甲烷和二氧化碳的效率提升,有利于反应器产甲烷性能提高.

2.1.3 盐度对气体产量与组成的影响

从图 4 观察到气体产量和甲烷百分比在盐度为 $1.3 \sim 2.1 \text{ g·L}^{-1}$ 时都达到了最大值,甲烷百分比稳定在 $46\% \sim 65\%$ 范围内,这与 Song 等 [18] 的研究结果相似;当盐度高于 2.1 g·L^{-1} 后,气体产量和甲烷占比整体呈下降趋势,在 110 d 后,与图 3(a) 和图



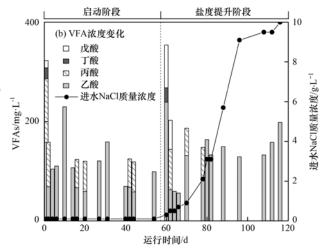


图 3 反应器出水 pH 和 VFA 浓度变化

Fig. 3 Variations of pH and VFA concentrations in permeate

3(b)对比发现虽然反应器内 pH 值升高,乙酸浓度和 VFA 总量增加,但气体产量和甲烷占比均明显下降,分析原因可能是一方面产酸菌对环境 pH 值的适应范围较宽,一些产酸菌在 pH 值为 5.5~8.5 范围内生长良好,另一方面产甲烷菌比产酸菌对盐度更敏感,高盐度使产甲烷菌细胞渗透压失衡、酶活性下降,且盐度越高乙酸硝化产生甲烷的量越少[19-23].

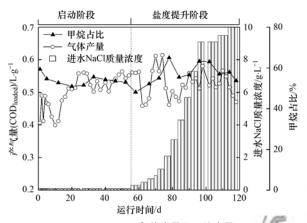


图 4 AnMBR 气体产量和甲烷含量

Fig. 4 Gas production and methane content in the AnMBR

2.2 AnMBR 污泥性质

2.2.1 盐度对污泥浓度的影响

图 5 反映了 AnMBR 运行期间污泥浓度的变化情况. 混合液悬浮固体浓度(MLSS)和混合液挥发性悬浮固体浓度(MLVSS)在启动阶段略有波动,基本稳定在 5.0~6.0 g·L⁻¹之间;随着盐度提升,MLVSS/MLSS 不断下降,二者均呈现先增加趋势,且在盐度为 2.1~9.1 g·L⁻¹范围内增加最明显,其原因可能是活性污泥中的微生物在这一盐度范围内生长代谢加快,微生物量增加;当盐度大于 9.1 g·L⁻¹时,二者均减少,说明过高的盐度会使微生物裂解死亡并分泌大量胞外聚合物最终使污泥浓度下降^[18,24].

2.2.2 盐度对污泥沉降性能的影响

分析图 6 发现污泥体积指数(SVI)在启动阶段由 126.94 mL·g⁻¹突增到 140.38 mL·g⁻¹,这可能是水力停留时间过短同时反应器运行初期存在跑泥现象造成的;随着反应器内有机负荷的增加,SVI逐渐降低,说明污泥絮体因微生物生长代谢所分泌的胞外产物而絮凝在一起,使得污泥沉降性能增强. 盐度提升阶段,SVI值在盐度为 0.3~3.1 g·L⁻¹范围内下降,说明这一范围的盐度使得各个微生物群落结合得更紧密;但当盐度继续升高时,SVI值增加,可能是高盐度微生物细胞裂解死亡后细胞质和分泌物大量悬浮在混合液中,导致污泥沉降性能变差.

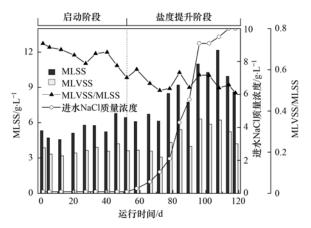


图 5 活性污泥 MLSS 和 MLVSS 变化

Fig. 5 Variations of MLVSS and MLVSS

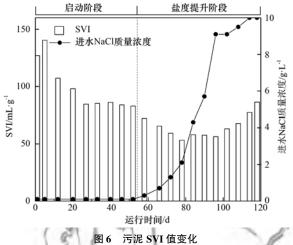


图 6 污泥 SVI 恒受化 Fig. 6 Variations of SVI

2.2.3 盐度对 SMP、EPS 的影响

图 7 直观地呈现了盐度提升阶段 AnMBR 污泥混合液中溶解性微生物产物(SMP)和胞外聚合物(EPS)的变化. SMP[图 7(a)]随盐度提升先稳中有降后增加,除个别点外 EPS[图 7(b)]随盐度提升而增加. 分析原因可能是一方面一定范围盐度促进微生物生长代谢提高了其对多糖、蛋白质等有机物的利用能力,分泌更多的胞外聚合物;另一方面,过高的盐度使微生物细胞脱水裂解死亡释放出蛋白酶等有机物,导致污泥混合液中蛋白质和多糖的积累^[25,26].

2.3 膜污染特性

2.3.1 膜污染程度表征

跨膜压差(TMP)是衡量膜污染程度的重要参数,这是因为与膜结垢有关的颗粒和胶体等物质会堵塞膜孔导致跨膜压差明显增加^[27]. 以膜组件开始运行到 TMP 超过 35 kPa 取出清洗计为一个周期,分析图 8 发现 AnMBR 的中空纤维膜组件在 118 d内一共运行了 3 个周期,启动阶段 TMP 的快速增加可能是由于膜孔的快速阻塞和胶体物质在膜表面

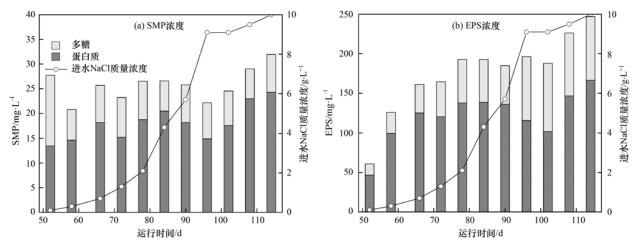


图 7 SMP 和 EPS 浓度变化

Fig. 7 Variations of SMP and EPS concentrations

聚集形成凝胶层^[28]. 随后膜的运行周期延长,这可能是因为反应系统运行稳定,适当增加盐度能促进微生物对有机物的降解效能,污泥中的 SMP、EPS中的蛋白质和糖类等物质减少,膜污染相较第 1 周期减轻^[24]. 而膜运行的第 3 周期 TMP 较前两周期增加得更为缓慢,除以上原因外还可能是反应器长期在恒定过滤时间和恒定过滤流量的条件下运行,凝胶层和滤饼层积累得更为缓慢^[16].

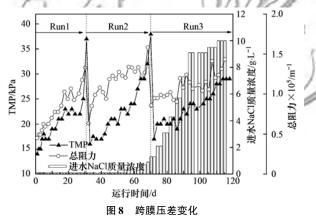


Fig. 8 Variations of membrane TMP

膜总阻力的变化规律与 TMP 基本相同. 使用物理化学法清洗被污染的膜组件, 重新运行时起始的膜总阻力总是要高于前一周期, 分析其原因可能是物理化学法并不能清洗掉全部的膜垢, 一些难以降解的物质会不断积累在膜孔中形成不可逆膜垢.

2.3.2 膜面污染物的扫描电镜-能谱(SEM-EDX)分析

通过 SEM 观察运行结束后膜面污染物的形态

(图9),发现大量颗粒、胶体物质聚集在中空纤维膜表面形成密实的污染层^[28],几乎观察不到未被堵塞的膜孔,说明膜污染十分严重.膜面污染物中还出现一些方形的、有棱角的、类似结晶状的物质,这极有可能是污泥混合液中的无机盐离子在膜表面结晶所致^[29].

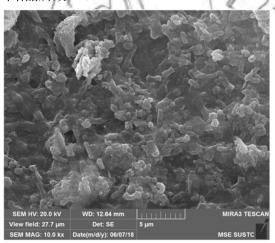


图 9 膜面污染物 SEM 图

Fig. 9 SEM image of membrane foulants

采用 EDX 分析膜面污染物中的无机成分组成发现(图 10 和表 1), C、O、S 可认为是膜面污染物的有机成分, 无机成分则为 Na、Mg、Al、Si、Cl、K、Ca 和 Fe. Wang 等^[30]发现由 Mg、Al、Fe、Ca 和 Si等元素组成的无机物质沉积在膜表面和有机物结合形成的凝胶层是造成膜污染的重要原因. 金属离子在通过膜时,可被微生物细胞和大分子有机物因电荷中和而络合,形成致密的膜垢,加速膜污染^[31].

表 1 膜面污染物中主要元素所占质量分数/%

Table 1	Quality score	of major element	s involved ir	n the memb	orane fouling/%	
N.	3.6	4.1	G:	C	CI	

元素	С	0	Na	Mg	Al	Si	S	Cl	K	Ca	Fe
质量分数	54. 25	33. 62	0. 97	0.71	1. 43	3. 14	1. 82	0. 26	0. 33	0. 44	3. 04

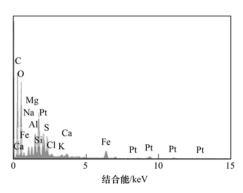


图 10 膜面污染物 EDX 图

Fig. 10 EDX results for the membrane foulants

2.3.3 膜面污染物的有机成分分析

目前普遍认为溶解性微生物产物(SMP)和胞外聚合物(EPS)是膜面污染物主要有机成分,其组成包括糖类和蛋白质等^[32].表2展示了污泥混合液和膜污染物中SMP、EPS的含量,其中膜污染物中两者的含量要远大于混合液中的含量,分析原因可能是混合液中的污染物在反应器长期运行中不断在膜面积累,并通过絮凝、络合、架桥等一系列相互作用,使得污染物之间结合更加致密^[24].

表 2 污泥混合液与膜面污染物中 SMP 和 EPS 含量(MLSS)/mg·g⁻¹

Table 2 SMP and EPS contents of mixed liquid and

membrane fouling(MLSS)/mg \cdot g $^{-1}$ 污泥混合液 膜污染物 样品 SMP SMP **EPS** 5.8 ± 1.4 39.6 ± 0.7 15.5 ± 1.3 147.5 ± 1.4 多糖 1.8 ± 0.9 19. 1 ± 1.2 6.1 ± 0.8 58.9 ± 1.6

图 11 显示了通过三维荧光光谱(EEM)分析膜面污染物的结果. 观察到图谱中主要有 3 个荧光峰,对应的有机成分分别为中等激发波长类色氨酸(IV区)、低激发波长类络氨酸(II区)以及高激发波长类腐殖酸(V区)^[33]. 这表明蛋白质与腐殖酸类物质是膜面有机污染物的主要成分.

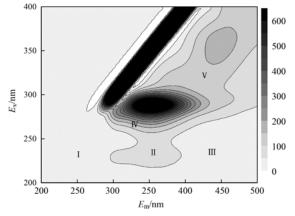


图 11 膜面污染物 EEM 图

Fig. 11 EEM results for the membrane foulants

3 结论

- (1)盐度对 AnMBR 处理废水的运行效能有较大影响,当盐度大于 9.5 g·L⁻¹时, COD 去除率迅速从 98%降至 80%以下,反应器内挥发性脂肪酸积累,气体产量和甲烷占比均减少.
- (2)随着 AnMBR 系统的盐度不断提升, 污泥性质有较大变化, MLSS 和 MLVSS 缓慢增长, 在盐度为 $9.1~{\rm g\cdot L^{-1}}$ 时达到最大值, SVI 先从 126. 94 mL·g⁻¹降至 53. 23 mL·g⁻¹后上升, SMP 和 EPS 增长相对稳定.
- (3)中空纤维膜组件在118 d 内运行了3 个周期,且随盐度增加膜运行周期由31 d 延长为48 d.用 SEM-EDX 分析发现膜面污染物中有类似结晶状物质,Na、Mg、Al、Si、Cl、K、Ca和Fe为主要无机元素.用 EEM 分析膜面污染物发现蛋白质与腐殖酸是膜面有机污染物的主要成分.
- (4)综合盐度对 AnMBR 运行特性、污泥性质和膜污染的影响,将进水盐度逐渐提升并控制在2.1~3.1 g·L⁻¹范围内能使系统处理含盐有机废水效率达到最高,并有效减缓膜污染.

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