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电导率对厌氧产酸、正渗透与微生物燃料电池耦合工艺运行性能的影响

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摘要:将厌氧产酸(AA)、正渗透技术(FO)与微生物燃料电池(MFC)进行耦合,构建了用于污水处理的 AAFO-MFC 耦合工艺,实现污水的同步产电和回用.由于电导率是 AAFO-MFC 运行的关键因素,考察了电导率对系统运行性能的影响.结果表明,较高的电导率可以降低 MFC 的内阻,提高产电,但是会加重 FO 膜污染,导致 FO 膜通量快速衰减,缩短运行时间.电导率对出水水质并没有显著影响,FO 膜出水的总有机碳(TOC)和总磷(TP)浓度分别低于 4 $\operatorname{mg·L^{-1}}$ 和 0.5 $\operatorname{mg·L^{-1}}$,但是 FO 膜对于氦氮(NH $_4^+$ -N)的截留效果较差.控制反应器内电导率为 7~8 $\operatorname{mS·cm^{-1}}$ 时,系统整体性能表现最佳,可以获得连续且相对稳定的输出电压,而且 FO 膜通量下降较为缓慢,运行周期达到 29 d.

关键词:电导率;微生物燃料电池;正渗透;产电;污水处理

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Effects of Conductivity on Performance of a Combined System of Anaerobic Acidification, Forward Osmosis, and a Microbial Fuel Cell

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Abstract: In this study, a novel combined system for simultaneous recovery of bioelectricity and water from wastewater was developed by integrating anaerobic acidification and a forward osmosis (FO) membrane with a microbial fuel cell (AAFO-MFC). Conductivity was thought to be an important factor affecting the performance of the AAFO-MFC system. Thus, effects of conductivity on the performance of AAFO-MFC system in treating synthetic wastewater were investigated. The results indicated that a higher conductivity increased the bioelectricity production, owing to a reduction in the internal resistance. However, it resulted in a rapid decrease of FO water flux and a shorter operating time because of a severer membrane fouling. The conductivity had no impact on the water quality of the effluents. The total organic carbon (TOC) and total phosphorus (TP) concentrations in the FO permeate were less than 4 and 0.5 mg·L⁻¹, respectively, at all conductivity levels. However, the rejection of the FO membrane for NH₄⁺-N was lower at all conductivity levels. The optimal comprehensive performance of this system was obtained when the conductivity was maintained at 7-8 mS·cm⁻¹. In this case, the AAFO-MFC system achieved continuous and relatively stable power generation, and the water flux of FO membrane was relatively stable during a long-term operation of approximately 29 days.

Key words: conductivity; microbial fuel cell; forward osmosis; bioelectricity production; wastewater treatment

正渗透(FO)和微生物燃料电池(MFC)作为两种新兴的技术,已经在污水处理与回用领域引起越来越多的关注. FO 过程利用膜两侧的渗透压差作为驱动力,使水分子自发地通过半透膜,具有出水水质好和膜污染趋势小等优点[1-3]. 然而,采用 FO 处理污水时,污水中所含的有机物仅仅是被截留,并未得到回收利用[4]. MFC 利用微生物作为催化剂,可将污水中的有机物直接转化成电能,但其出水水质较差,无法直接排放或回用[5-8]. 针对 FO 和 MFC 的优点和缺点,有学者提出将两种技术进行耦合,在同一系统中同时实现污水回用和生物电回收[9,10].目前, FO 与 MFC 的耦合方式主要是采用 FO 膜代

替 MFC 中的质子交换膜,形成正渗透微生物燃料电池(OsMFC),借助 FO 膜的高效截留能力提高出水水质和产电性能^[9~13]. 然而, FO 膜的反向盐渗透和膜污染限制了 OsMFC 的长期稳定运行^[14~16]. 针对 OsMFC 存在的问题, Liu 等^[17]将 FO 膜、厌氧产酸(AA)和 MFC 进行全新的耦合(AAFO-MFC),改变 FO 膜作为质子交换膜的作用,利用其对小分子有机物的高效截留能力,实现污水水质的提升以及

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有机物的富集,然后利用 MFC 使有机物以生物电的 形式回收,实现了系统的长期稳定运行.

在 AAFO-MFC 系统中,较高的电导率不仅导致 FO 膜通量快速下降,而且影响污泥浓度、污泥粒径 以及胞外聚合物(EPS)等污泥性质,加重 FO 膜污染^[16]. 然而,在一定范围内提高电导率可以促进离子传递,从而降低内阻,提高 MFC 的产电性能^[14,15]. 因此,电导率是限制 AAFO-MFC 系统运行的关键因素,寻找最佳的电导率具有十分重要的意义. 本研究的目的就是考察电导率对 AAFO-MFC 系统运行性能的影响,以期获得最佳的电导率.

1 材料与方法

1.1 实验装置

AAFO-MFC 系统由生物反应器和单室空气阴极 MFC 两部分耦合而成,如图 1 所示. 生物反应器的 有效体积为 4.5 L,内置 FO 和微滤(MF)两个膜组 件. FO 膜的材质为醋酸纤维(CTA),有效膜面积为 0.025 m²,采用活性层朝向原料液(AL-FS)的方式 运行. FO 膜采用 0.5 mol·L⁻¹的 NaCl 作为汲取液. 为了维持汲取液盐度的稳定,借助电导率控制仪,通过补充 5 mol·L⁻¹的高浓度 NaCl 溶液来实现. MF 膜为聚偏氟乙烯(PVDF)材质,平均孔径为 0.2 μm,有效膜面积为 0.025 m². 为了减缓膜污染以及提高传质效果,借助自身产生的生物气进行曝气. 气体循环速率控制在 2 L·min⁻¹左右. 为了提高产酸的效果,通过向反应器中加入 NaOH 溶液来维持反应器中污泥混合液的 pH 在 9.5 左右.

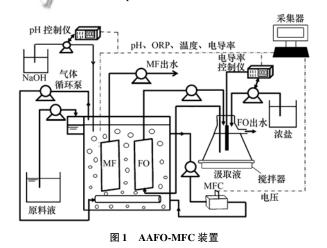


Fig. 1 Flow diagram of the AAFO-MFC system

本实验采用单室空气阴极 MFC,其有效体积为 160 mL. MFC 与生物反应器联通,即生物反应器中的污泥混合液进入 MFC,在产电菌利用厌氧产酸产

生的小分子有机物之后,再回流到生物反应器中. 阳极材料为石墨毡(三业碳素有限公司,北京),电极面积 $40~{\rm cm}^2$. 阴极为活性炭-PTFE 空气阴极,电极面积为 $40~{\rm cm}^2$. MFC 两电极间通过外接 $500~\Omega$ 的电阻相连接.

1.2 运行条件

AAFO-MFC 的进水为模拟生活污水,其总有机碳(TOC)、氨氮(NH₄⁺-N)、总氮(TN)和总磷(TP)浓度分别为(146.3±2.69)、(28.1±1.19)、(31.7±1.90)和(2.2±0.11)mg·L⁻¹.模拟生活污水的配方为:葡萄糖 230 mg·L⁻¹、蛋白胨 60 mg·L⁻¹、无水乙酸钠 40 mg·L⁻¹、牛肉膏 20 mg·L⁻¹、NaHCO₃ 198 mg·L⁻¹、KH₂PO₃ 12 mg·L⁻¹、NH₄HCO₃ 170 mg·L⁻¹、MgCl₂·6H₂O 2.4 mg·L⁻¹、无水 CaCl₂ 1.2 mg·L⁻¹、FeCl₃·6H₂O 1 mg·L⁻¹、接种污泥取自无锡太湖新城污水处理厂,在运行前首先用模拟生活污水在碱性条件下进行厌氧驯化.待厌氧酸化污泥性质稳定后,再接种到生物反应器中、MFC 直接利用生物反应器中的混合液作为接种物和底物,直至连续稳定产电,完成驯化过程.

为了避免高盐度带来的不利影响,一般控制耦合 FO 的生物反应器的电导率为 4~5 mS·cm^{-1[17]}. 此外,根据 Fan 等的研究^[18],当电导率为 9~10 mS·cm⁻¹时,产电效果较好. 基于以上分析,为了考察电导率对 AAFO-MFC 整体运行性能的影响,通过调节 MF 膜的通量,分别将生物反应器中的电导率控制在 4~5、7~8 和 9~10 mS·cm⁻¹.

整个实验过程都在恒温室中进行,温度始终维持在(28 ± 0.5) °C. 污泥停留时间(SRT) 控制在 80 d,混合液悬浮物浓度(MLSS) 为 2 ~ 3 g·L⁻¹. 为了获得较好的产酸效果,在实验过程中使用 0.2 mol·L⁻¹的氢氧化钠(pH = 13.5) 调节反应器中的pH 值在 9.5 \pm 0.2 [17, 19,20].

1.3 测定方法

1.3.1 水质分析

膜通量采用单位时间内通过单位膜面积的透过液量来表示,单位L·(m²·h)⁻¹. 电导率和 pH 分别采用便携式电导率仪(EC300A, YSI, 美国)和台式pH 计(DELTA 320,梅特勒-托利多)进行测定. 进水、滤液、MF 出水、FO 出水的氨氮、TN、TP 以及MLSS 和混合液挥发性悬浮固体浓度(MLVSS)均采用国家标准方法测定^[21]. 进水、滤液、MF 出水、FO 出水的 TOC 采用 TOC 分析仪(TOC-Vesh,岛津,日本)测定. 污泥混合液中 EPS 采用加热法提

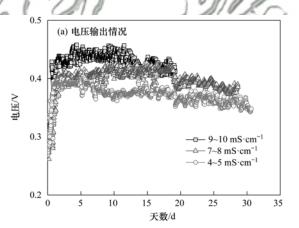
取^[22],其浓度采用蛋白质和多糖加和的形式表示. 蛋白质采用考马斯亮蓝法测定^[22],而多糖采用苯酚-硫酸法测定^[22].

1.3.2 电化学分析

输出电压 U 由数据采集卡(RBH8221 型,瑞博华,北京)自动记录,每 30 min 记录一次. 电流 I 由欧姆定律 I=U/R 计算,其中 R 为外电阻. 电压输出达到峰值且稳定时,通过改变外电阻(10~10000 Ω)分别测得 MFC 在不同外电阻时的输出电压,然后根据 $P=U^2/R$ 、p=IU/A 和 i=I/A,分别求得输出功率、功率密度和电流密度,并绘制极化曲线和功率密度曲线. MFC 内阻由极化曲线的斜率计算得到[23].

1.3.3 VFA 的测定

采用 GC-2010 气相色谱仪(岛津,日本)测定污泥混合液中的挥发性脂肪酸(VFA). 将 2 mL 污泥混合液在12 000 r·min $^{-1}$ 下离心 10 min,所得上清液经 0. 45 μm 膜过滤后,取 0. 5 mL 滤液于离心管中,加入等体积 3 mol·L $^{-1}$ 的磷酸酸化后,再加入等体积 0. 835 g·L $^{-1}$ 的 4-甲基戊酸作为内标物进行气相色谱分析. 采用 AOC-20i 自动进样装置. 毛细管柱型号 PEG-20M, 30 m×0. 32 mm×0. 50 μm. 气化室温度 250 $^{\circ}$ C. 采用一阶程序



升温,柱箱初始温度 80℃,保留 2 min,升温速率 15 $℃ \cdot min^{-1}$,升至 190℃,停留 1 min. 氮气作为载气.

2 结果与讨论

2.1 电导率对产电的影响

不同电导率条件下 AAFO-MFC 电压输出情况 如图 2 所示. 从中可以看出,在每个电导率条件下, AAFO-MFC 系统均表现出连续且相对稳定的电压输 出,但随着运行时间的增加,电压均略有下降并分别 稳定在 0.41、0.39 和 0.35 V. 整个运行周期结束 后,通过改变外阻,获得 MFC 的极化曲线和最大功 率密度曲线. 由图2可知,电导率为9~10 mS·cm⁻¹ 时, MFC 获得的最大功率密度最高, 为 4.55 W·m⁻³, 电导率为7~8 mS·cm⁻¹时次之, 而电导率 为4~5 mS·cm⁻¹最低. 与电导率为9~10 mS·cm⁻¹ 相比,电导率为7~8 mS·cm⁻¹和4~5 mS·cm⁻¹时, MFC 所获得的最大功率密度分别下降了 7.4% 和 22.2%. 各电导率所对应 MFC 的表观内阻分别为 167.1、181.4 和 234.8 Ω. 以上结果表明, 电导率越 高, MFC 内阻越低, 获得的功率密度也越大. 这是 由于较高的电导率会促进电子以及离子传递,从而 降低内阻,提高产电性能[15,24].

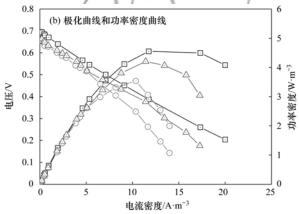


图 2 不同电导率条件下电压输出情况以及极化曲线和功率密度曲线 Fig. 2 Electricity generation, power density, and polarization curves at different conductivities

2.2 电导率对 FO 膜通量的影响

不同电导率条件下, FO 膜通量随时间的变化如图 3 所示. 从中可以发现,在所有电导率条件下, FO 膜通量都分为初期的快速下降和后期的缓慢下降两个阶段. 当 FO 膜通量降至 2 L·(m²·h)⁻¹以下时, AAFO-MFC 运行结束. 这与相关报道中 FO 膜处理污水过程中的通量变化情况一致^[25].

此外,从图 3 还可以看出,在电导率为 9~10 mS·cm⁻¹时, FO 膜通量下降较快,仅在运行 19 d

后,通量即降至 $2 \text{ L·}(\text{m}^2 \cdot \text{h})^{-1}$ 以下,通量下降速率为 $0.25 \text{ L·}(\text{m}^2 \cdot \text{h})^{-1} \cdot \text{d}^{-1}$. 然而,在电导率为 $7 \sim 8$ mS·cm⁻¹和 $4 \sim 5$ mS·cm⁻¹时, FO 膜通量的下降则较为平缓,运行周期分别为 29 d 和 31 d,对应的通量下降速率分别为 $0.15 \text{ L·}(\text{m}^2 \cdot \text{h})^{-1} \cdot \text{d}^{-1}$ 和 $0.14 \text{ L·}(\text{m}^2 \cdot \text{h})^{-1} \cdot \text{d}^{-1}$. 与电导率为 $9 \sim 10 \text{ mS·cm}^{-1}$ 相比,电导率为 $7 \sim 8 \text{ mS·cm}^{-1}$ 和 $4 \sim 5 \text{ mS·cm}^{-1}$ 时, AAFO-MFC 系统的运行时间分别提高了 52.6% 和 63.1%. 因此,较高的电导率水平不利于反应器长

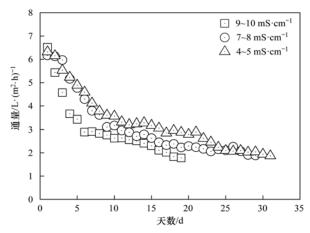


图 3 不同电导率条件下膜通量随时间的变化

Fig. 3 Changes in water fluxes of the FO membrane with operating time at different conductivities

期运行. 然而,相比于 7~8 mS·cm⁻¹,较低的电导率水平(4~5 mS·cm⁻¹)并没有明显延长 FO 膜运行时间,这可能是由于电导率为 4~5 mS·cm⁻¹时, FO 膜通量较大,通量的增加反而会造成更加严重的膜污染^[26].

2.3 电导率对出水水质的影响

2.3.1 TOC 去除以及 VFA 变化

为了考察电导率对 AAFO-MFC 系统去除有机物的影响,分别定期测定进水、滤液、MF 出水及FO 出水中 TOC 的变化,结果如图 4 所示. 从中可以看出,在各个电导率条件下, AAFO-MFC 系统对有机物都表现出较好的整体去除效果. 其中,由于 FO 膜对有机物具有较好的截留性能,每个电导率条件下的 FO 出水中 TOC 的浓度均低于 4 mg·L⁻¹, FO 膜对 TOC 的去除率均大于 97%. 相对于 FO 膜,MF 膜出水中的 TOC 浓度略高,这表明 FO 膜截留能力优于 MF 膜. 相比之下,控制反应器电导率为 9~10 mS·cm⁻¹的条件下,滤液中的 TOC 浓度较其他电导率条件明显要高,这可能是由于高的电导率条件对微生物活性有一定的抑制作用,影响了生物反应器对有机物的去除.

此外,在不同电导率条件下,污泥混合液中的VFA 仅有乙醇被检测到,并未发现其他的挥发酸,比如乙酸等.相关文献表明,乙醇同样可以作为MFC 的底物^[27,28].这说明,在 AAFO-MFC 系统中,主要通过厌氧产酸过程将复杂有机物转化为易被MFC 利用的小分子乙醇和有机酸,同时利用 FO 膜的高效截留能力实现积累,从而提高产电.

2.3.2 NH₄ -N、TN 和 TP 的变化

由图 5 可以发现, AAFO-MFC 系统在不同电导

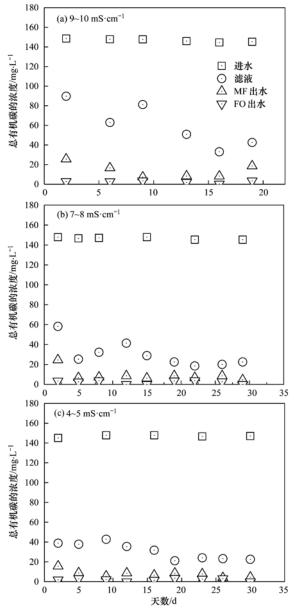


图 4 不同电导率条件下进水、滤液、MF 出水和 FO 出水中 TOC 的变化

Fig. 4 Variations in TOC concentrations of the influent, sludge supernatant, and MF and FO effluents at different conductivities

率条件下运行时,FO 膜对NH₄⁺-N的截留效果均不太理想,出水中NH₄⁺-N浓度仍然较高,对NH₄⁺-N的截留率分别为 20% ~ 28%,16% ~ 23% 和 18% ~ 25%. 事实上,FO 膜对NH₄⁺-N截留效果差这一现象在文献[15]中也有报道. 相比于 FO 膜,MF 膜截留能力更差,甚至对NH₄⁺-N没有截留. 此外,由于生物反应器处于厌氧酸化阶段,主要发生有机氮向NH₄⁺-N的转化,NH₄⁺-N就成为 TN 的主要存在形式. 因此,滤液、MF 和 FO 出水中 TN 的变化规律与NH₄⁺-N相同,不再赘述. 对于 TP 来说,由图 6 可以看出,在不同电导率条件下反应器中的 TP 均出现

一定程度的累积,但由于 FO 膜的高效截留作用, FO 出水中的 TP 浓度均低于 $0.5 \text{ mg} \cdot \text{L}^{-1}$,对 TP 的

截留率均达到 95% 以上, 明显优于 MF 膜. 电导率 并未对NH⁺-N和 TP 的去除产生显著影响.

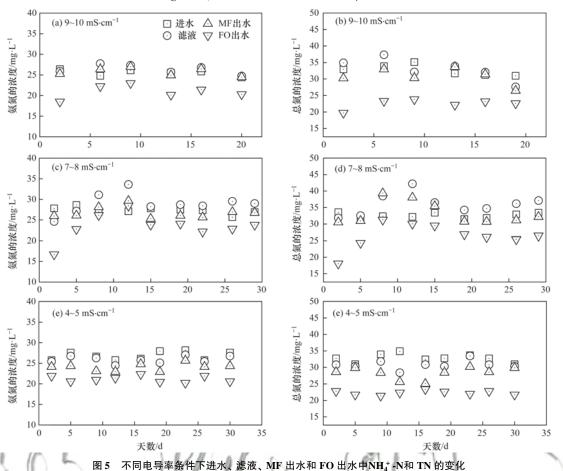


Fig. 5 Variations in NH₄⁺-N and TN concentrations of the influent, sludge supernatant, and MF and FO effluents at different conductivities

2.4 EPS 变化情况

EPS 主要由微生物产物(SMP)和胞外聚合物 (BEPS)两部分组成,对污泥性质以及膜污染均具有 较大的影响. 在本实验中,进水主要以葡萄糖作为 碳源,因此, SMP 和 BEPS 均以多糖为主^[22]. 在不 同电导率条件下,反应器中SMP、BEPS以及污泥浓 度的变化情况如图 7 所示. 从中可以看出,当电导 率为9~10 mS·cm⁻¹时,污泥浓度明显下降,从最初 的 2.54 g·L⁻¹左右下降至 1.08 g·L⁻¹左右,而其他 两组实验中污泥浓度虽有所降低,但下降幅度较小. 导致污泥浓度下降的原因主要有两方面,一方面是 由于通量的下降,水力停留时间过长,营养物质匮 乏,细胞发生内源消化[29];另一方面,反应器内盐 度的累积对微生物的活性产生抑制[28]. 对比3组 实验可知,后者是导致本实验中污泥浓度下降较快 的主要原因. 此外,反应器内电导率为9~10 mS·cm⁻¹时, SMP 和 BEPS 的浓度均明显高于其他 两组实验. 这是因为在较高的盐度环境下,微生物 细胞为了适应环境会发生水解等现象,导致 EPS 含量会有所上升^[2]. 考虑到 EPS 对膜污染的重要贡献,较高的 EPS 含量可能会加重 AAFO-MFC 系统运行过程中 FO 膜的生物污染. 因此,控制反应器内电导率为 9~10 mS·cm⁻¹会造成较为严重的膜污染,缩短 FO 膜运行时间,这与 2.2 节中的实验结果相一致.

3 结论

- (1) 较高的电导率可以降低 MFC 的内阻,提高产电. 然而,较高的电导率对微生物活性会产生抑制,造成 EPS 浓度的上升,加重膜污染,导致 FO 膜通量快速衰减,缩短运行时间.
- (2) 电导率对出水水质没有显著影响. 在各电导率条件下, FO 膜对有机物以及 TP 均具有较好的截留能力, 其中 FO 出水中有机物去除率均可达97%以上, FO 出水中 TP 均低于 0.5 mg·L⁻¹. 然而, FO 膜对于NH₄⁺-N的截留效果较差.

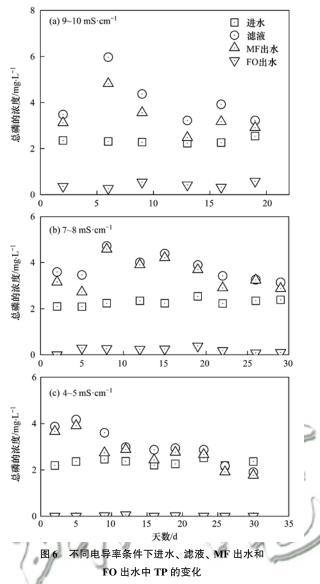
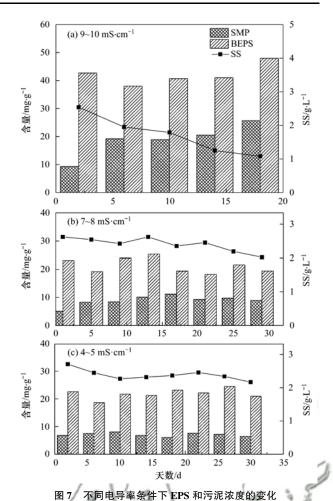


Fig. 6 Variations in TP concentrations of the influent, sludge supernatant, and MF and FO effluents at different conductivities

(3) AAFO-MFC 的最佳电导率为 7 ~ 8 $mS \cdot cm^{-1}$,此时污泥性质较为稳定,FO 膜通量衰减相对较小,电压稳定在 0.39 V,最大功率密度为 4.21 $W \cdot m^{-3}$.

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