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STUDY ON RELIEVING BIO-CLOGGING WITH THE HYBRID SYSTEM OF CONSTRUCTED WETLAND AND MICROBIAL FUEL CELL

LI Ang, AI Peng, LIN Li-Li, LU Rui, RONG Peng, LI Zhi-Wei, XIAO En-Rong, WU Zhen-Bin

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人工湿地-微生物燃料电池耦合系统缓解生物堵塞试验研究

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摘要: 微生物燃料电池(Microbial Fuel Cell, MFC)利用电极表面富集的电化学活性菌(Electrochemically Active Bacteria, EAB)对生物堵塞的胞外聚合物(Extracellular Polymeric Substances, EPS)进行降解, 并通过产电过程中形成的微弱电场来抑制微生物胞外多糖大量分泌, 可在一定程度上控制人工湿地(Constructed Wetland, CW)的生物堵塞。为此, 研究将阴极和阳极电极分别嵌入垂直流人工湿地的不同深度位置处, 构建了人工湿地-微生物燃料电池(CW-MFC)耦合系统。通过比较开路和闭路耦合系统的孔隙率、过水速率以及净化效果, 评价CW-MFC系统的堵塞延缓能力。结果表明: 相较开路系统, 闭路系统的过滤速率增幅较大, 孔隙率的降幅较小, 在一定程度上缓解了堵塞。闭路系统对TN和NH_4^+-N的去除率显著高于开路系统。嵌入电极形成的CW-MFC系统可原位缓解堵塞, 有较好的应用潜力。

关键词: 人工湿地; 生物堵塞; 孔隙率; 过滤速率; 微生物燃料电池

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人工湿地(CW)堵塞是指运行过程中基质床渗透系数降低, 表面雍水, 污水处理性能下降的现象[1]。在高有机物条件下, 异养微生物大量分泌的胞外聚合物(Extracellular Polymeric Substances, EPS)过量积聚在基质表面并吸附大量的有机和无机物质而形成的生物堵塞, 会对人工湿地的净化效果和寿命造成更严重的影响[2—4]。CW中的生物堵塞相较于水悬浮物引起的物理堵塞、填料孔隙间发生化学反应产生胶体或沉淀引起的化学堵塞更为普遍, 是CW长效运行的主要限制因素[5, 6]。微生物燃料电池(Microbial Fuel Cell, MFC)可利用电极表面富集的电化学活性菌(Electrochemically Active Bacteria, EAB)对大分子有机物EPS的降解, 并通过产电过程中形成的微弱电场来抑制微生物胞外多糖的大量分泌, 一定程度上控制生物堵塞[7—9]。微弱电场可以刺激微生物生长, 促进污染物降解, 适当的电刺激有助于EPS分泌和生物膜形成[9]。EAB在阳极上富集可以增大功率输出, 从而为阴极去除污染物提供更多质子, 提高废水的处理效率[10]。此外, 电场可以防止污染物附着在膜上, 从而延缓膜污染[11]。MFC电极富集的EAB可将胞外聚合物中难降解的大分子有机物分解成小分子的、可溶性的有机物加以利用并产生电能[12]。大量研究[13—17]表明, MFC产生的微电场可以降低EPS含量并改变其有机物组成。MFC和膜生物反应器(MBR)结合可以减少污泥EPS含量, 减缓膜污染, 同时, MFC产生的电能可以减少系统电力输入, 降低能耗[18]。此外, EPS的主要组分腐殖质、蛋白及多糖类物质均为带负电荷的微粒, MFC形成的微弱电场可通过静电斥力作用促使负电荷微粒脱离聚合物表面并相互分离, 使得EPS组分进一步分散到液相, 进而更易于微生物降解利用。借助于CW垂直方向上天然具备的氧化还原电位梯度, 通过嵌入电极的方式, 可形成人工湿地-微生物燃料电池(CW-MFC)耦合系统[19]。该系统对于大量有机质引起的生物堵塞的缓解效果是本研究关注的重点。

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1　材料与方法

1.1　试验装置

试验装置如图 1所示, 为垂直流人工湿地柱形模拟装置, 直径100 mm, 高500 mm。反应器从底部往上依次填充: 100 mm厚的砾石层(粒径8—12 mm), 150 mm厚的颗粒活性炭阳极层(粒径3—5 mm), 200 mm厚的砾石层(粒径4—8 mm)。6 mm厚的石墨毡阴极层(直径为90 mm)。阳极层中插入卷成圆筒状的石墨毡(长×宽=200 m×150 m, 有效面积0.06 m^2)作为电流收集器。阳极和阴极采用直径1 mm的钛丝与1000 Ω的外电阻连接形成闭合回路。

1.2　试验设计

试验设计开路和闭路两组, 每组3个平行, 共6套装置。为了减少干扰, 本试验中不种植植物, 装置置于室内, 控制环境温度在25—30℃。接种污泥取自武汉市某污水处理厂好氧和厌氧反应池。首先将取回的活性污泥厌氧培养1周, 然后按30% (*v*/*v*)与配置的营养液充分混合后加入装置中进行挂膜。阳极石墨毡先浸泡在好氧活性污泥中48h进行微生物挂膜后再放入反应器的阳极层。整个接种驯化过程持续约1个月, 每周更换装置内接种液。当连续两周输出电压变化幅度均在10%以内, 出水COD去除率达到稳定时, 即可认定驯化完成。

试验分为模拟堵塞和缓解堵塞两个阶段。模拟堵塞阶段在开路状态下进行, 大约持续了1个月。将取自污水处理厂的剩余污泥在4℃下沉降浓缩24h, 再用自来水稀释以达到所需浓度。人工湿地堵塞物在基质间积聚的速度随着堵塞的发生而逐渐加快, 为更好的模拟人工湿地生物堵塞的发展过程, 实验设计投加的污泥浓度依次增加, 分别为30%、40%、50%、60%和70%, 每种污泥浓度投加2次, 每隔3天放水加污泥。当基质表面有明显雍水现象, 系统的孔隙率和过滤速率降低直到不再发生明显变化时, 视为系统已发生堵塞。缓解堵塞阶段, 闭路组连接1000 Ω外阻, 通过电压采集器实时监测电压。进水为人工配置的生活污水, 主要成份为乙酸钠、氯化铵和磷酸二氢钾。配置的进水中控制TN=30 mg/L, TP=2.5 mg/L左右, 以醋酸钠(CH_3COONa)作为COD来源, 分别控制为大约500和1000 mg/L两个浓度。每种进水浓度重复5次, 采用间歇方式运行1周。比较不同进水COD浓度下系统的产电性能、污水净化效果和湿地堵塞程度。

1.3　指标测定

堵塞程度用孔隙率和过滤速率表征。孔隙率采用排水法测定, 将试验装置通入自来水, 基质吸水达到饱和后, 量取充满水时(水面刚好没到基质表面)的填料高度H, 测定试验柱子的内径为D, 再把水放空, 测得放出水的体积V, 根据公式(1)计算孔隙率A。过滤速率则以单位时间内通过湿地填料的流量来计算, 为更好的进行对比, 对该测试方法进行了改良, 测定出水体积达到固定值V_0所用的时间t, 根据公式(2)计算过滤速率μ。

$$A = \frac{4V}{\pi D^2 H} \tag{1}$$

$$\mu = \frac{V_0}{t} \tag{2}$$

电压通过数据采集器(型号R6016/U/C3, 上海继升电气有限公司)每10s自动记录一次。分析数据时取10min内的平均值, 采集精度为0.001 V。极化曲线和功率密度曲线使用稳态放电法[20]测定。依次调减外阻从9000 Ω降到10 Ω, 记录相应的电压值, 采用采用极化曲线斜率法计算系统内阻R_{int} (为极化曲线斜率和阳极有效面积的比值)。

水质理化指标均参照国家环境保护标准执行: COD(HJ/T 399—2007)、TN(HJ 636—2012)、NH_4^+-N (HJ 535—2009)、NO_3^--N(HJ/T 346—2007)等的测定。

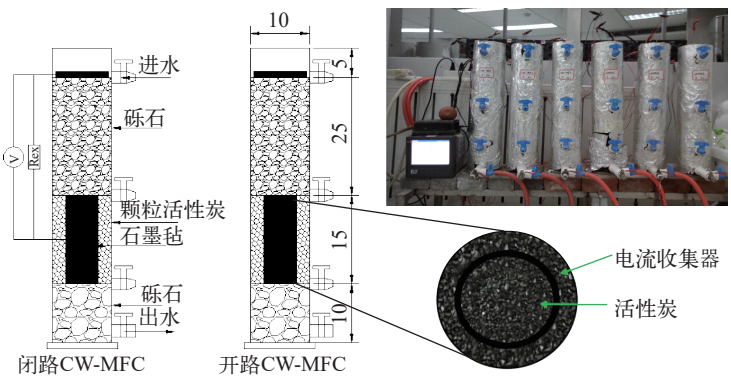

图 1　CW-MFC试验装置图

Fig. 1　Set up of CW-MFC systems

1.4 数据分析

试验数据使用SPSS 22.0 统计软件中的One-way ANOVA法进行单因素方差分析。

2 结果

2.1 系统堵塞程度

试验第一阶段系统堵塞之后, 闭路组和开路组系统堵塞后的平均孔隙率分别为(29.71±5.06)%和(26.31±1.66)%, 平均过滤速度分别为(0.758±0.152) L/min和(0.702±0.189) L/min。试验第二阶段堵塞缓解阶段的孔隙率和过滤速率的变化如图2所示。图2a显示, 闭路组和开路组的孔隙率很接近, 都在26%—30%小幅波动。尽管没有显著性差异(P>0.05), 但从趋势上看, 开路组的孔隙率在进水醋酸钠浓度为1000 mg/L时, 呈现出缓慢上升态势; 而闭路组则相对稳定。对于过滤速率指标而言, 尽管也无显著差异(P>0.05), 但在两种进水醋酸钠浓度下闭路组均高于开路组。由于开路组和闭路组的初始堵塞指标存在差异, 为了消除该影响, 分别以闭路和开路系统初始值(第0天)为参照, 计算每个实验周期的孔隙率和过滤速率的变化率。孔隙率的减少率的含义为: 当孔隙率的减少率为负值时表示孔隙率增加; 为正值时表示孔隙率减少。而过滤速率的增加率的含义为: 当过滤速率的增加率为正值时表示增加, 为负值时表示减少。如图2b所示, 闭路组的孔隙率减少率均为正值, 开路组则均为负值。这意味着相对于初始值, 闭路系统的堵塞情况在缓解, 而开路对照系统则呈现堵塞加重的趋势, 这在1000 mg/L的醋酸钠浓度时更为明显。再看过滤速率增加率指标, 高浓度时闭路系统显著高于开路系统, 也证明了对于堵塞有一定缓解作用。

2.2 产电性能

CW-MFC闭路系统的输出电压如图3所示。单个试验周期内输出电压呈先迅速飙升, 之后小幅稳定波动, 最后下降的趋势, 最大电压值均在400 mV上下, 并未随着进水COD浓度的倍增而增加, 其原因可能是MFC的产电量受多种因素影响, 除了底物浓度外, 装置构型、电极材料和电极间距等也会影响产电量, 因此其产电量不会无限增大, 即使底物浓度增加一倍, 其产电量也相对稳定。

两种进水浓度下的极化曲线和功率密度曲线趋势也相同(图4)。进水CH_3COONa=500 mg/L时开路电压(OCV)、最大功率密度(P_{max})和内阻(R_{int})分别为720 mV、2.85 mW/m^{2}和713 Ω。进水浓度达到1000 mg/L时, 相应的OCV、P_{max}和R_{int}分别为706 mV、3.08 mW/m^{2}和692 Ω。可见, 在两种浓度下上述参数值较为接近, 这与前述输出电压的结果一致。因为微生物的生长除了营养物质之外, 还受

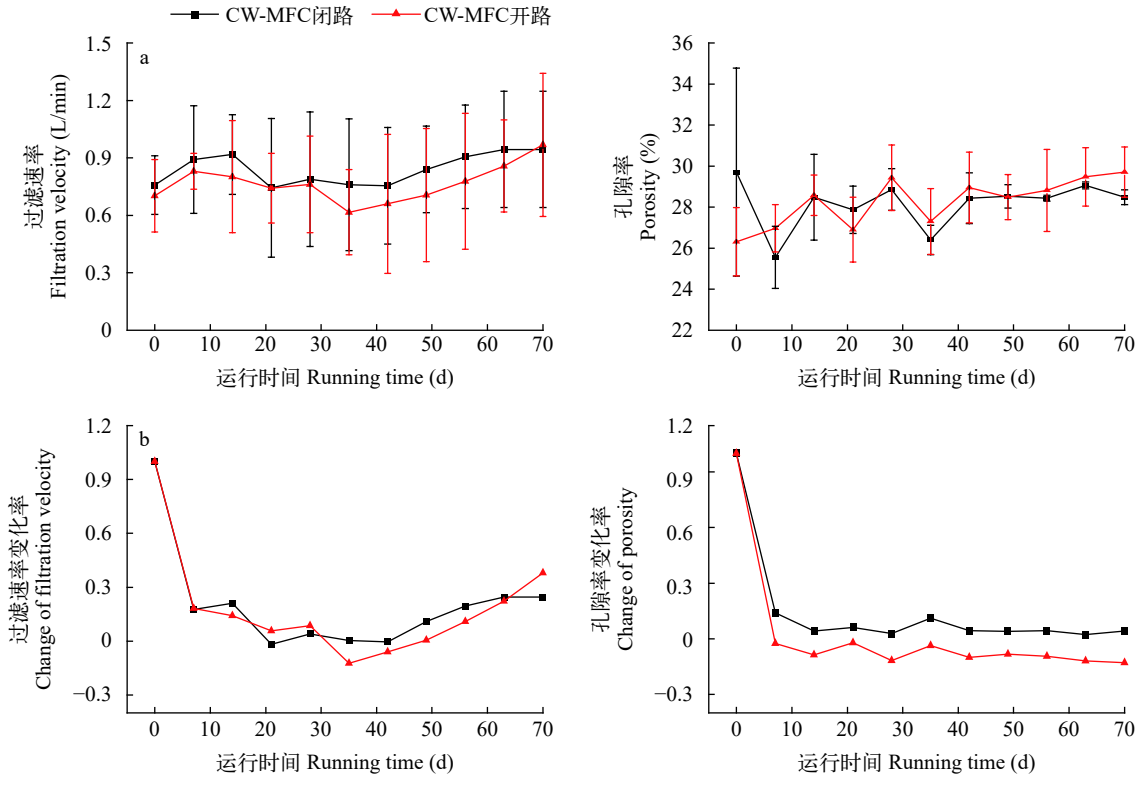

图2 闭路和开路CW-MFC系统的孔隙率和过滤速率(a)及其变化率(b)

Fig. 2 Porosity and filtration velocity (a) and their change rate (b) of CW-MFC systems with circuit and open modes

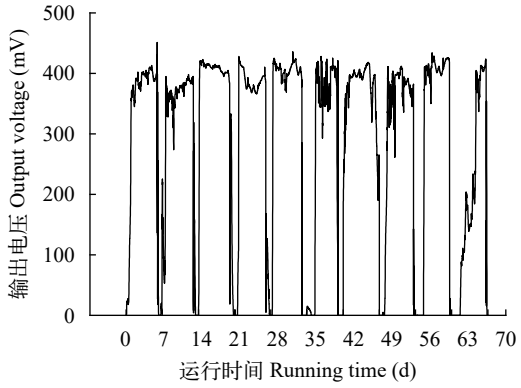

图 3　CW-MFC闭路系统输出电压

Fig. 3　Output voltage of the CW-MFC circuit system

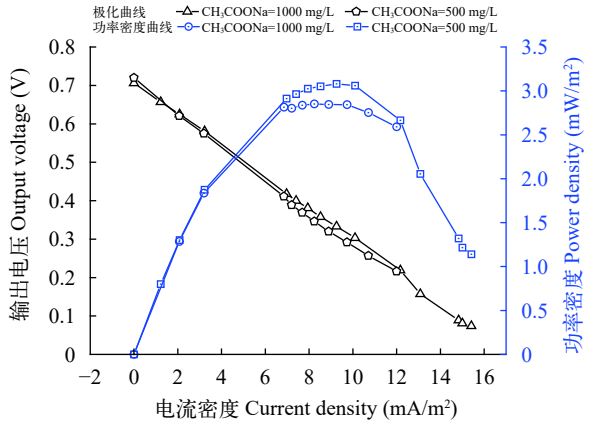

图 4　CW-MFC闭路系统极化曲线和功率密度曲线

Fig. 4　Polarization curves and power density curves of the CW-MFC circuit system

温度、pH、氧化还原电位、溶解氧等环境因子的影响, 不能无限生长; 所以两种浓度下的产电性能差异不大。

2.3　污水净化效果

系统的进出水理化指标pH、溶解氧(DO)、电导率(SPC)和氧化还原电位(ORP)如图 5所示。在两种浓度下, 闭路组和开路组出水中DO、pH、SPC、ORP的差别均不大, 闭路组基本上均略低于开路组。闭路组较进水有较大变化, DO均值从(3.92±0.35) mg/L降低到(1.37±0.35) mg/L, pH从7.41±0.23降低到6.68±0.08, ORP从(335.34±38.59) mV降到–60 mV以下。这些理化指标侧面说明阳极的电化学活性菌代谢有机物产生电能, 使得阳极酸化; 同时ORP的负值也说明了阳极室为厌缺氧环境。

图 6显示的是主要水质指标COD、TN和NH_4^+-N的浓度变化。进水CH_3COONa=500 mg/L时, 闭路系统出水COD、TN和NH_4^+-N的平均值分别为(34.62±7.39)、(8.68±1.42)、(8.36±1.74) mg/L; 开路系统出水COD、TN和NH_4^+-N的平均值分别为(30.35±9.34)、(14.87±1.63)、(13.59±0.60) mg/L。进水浓度为1000 mg/L时, 闭路系统出水COD、TN和NH_4^+-N的平均值分别为(28.55±4.28)、(7.43±1.63)、(6.29±0.85) mg/L; 开路系统出水COD、TN和NH_4^+-N的平均值分别为(28.65±5.90)、(13.43±1.32)、(11.19±1.27) mg/L。可见, 开路和闭路系统的去除效果在高浓度进水时均较好。

3　讨论

本研究中的堵塞缓解阶段, 当进水CH_3COONa浓度倍增时, 闭路系统COD平均去除率从(92.53±1.60)%增加到(96.93±0.46)%, 与开路系统几乎没有差别。但是闭路系统对TN和NH_4^+-N的平均去除率(均在70%以上)却显著高于开路系统(50%—60%)。进水中的TN以NH_4^+-N为主要形态(占比80%以上), 出水中仍以NH_4^+-N为主要形态, 占比达到90%以上。这说明系统中没有硝酸盐积累, 转化的NO_3^--N很快被还原成了N_2。其实进一步分析可知, 进水C/N远大于5, 所以系统内异养反硝化菌会大量繁殖, 不会出现NO_3^--N积累; 而随着进水COD倍增, 系统内趋向于缺氧环境(ORP减少), NH_4^+-N转化为NO_3^--N过程受阻, 所以TN和NH_4^+-N的去除率均下降。闭路系统中更高的TN和NH_4^+-N去除率(较开路系统高出30%—40%), 意味着产电环境有利于脱氮菌的富集。同时, COD去除率随着进水浓度增加略有上升, 闭路系统的输出电压却没有变化, 说明系统中阳极富集的电活性菌丰度没能进一步增加, 这可能与电活性菌与反硝化菌的竞争有关。此外, 在高浓度进水时, COD出水更低, 意味着更多的COD被降解利用, 而转化为电能的没有增加, 说明更多的用于了异养菌的生长。

EPS的主要来源有细胞的分泌物、脱落的细胞表面物质、细胞自溶物以及从环境中吸附的物质[21]。EPS既存在于细胞外, 也存在于微生物聚集体的内部。通常认为细胞的死亡和自溶将高分子量的化合物释放到介质中而形成EPS, 典型的如胞内储存的作为碳源和能源的聚β-羟基链烷酸酯或糖原, 还有完整的细胞成分如细胞壁和细胞膜(如磷脂)等[22]。当有机营养充足时, 异养微生物可大量分泌多糖、蛋白等物质到胞外, 形成EPS[23]。多糖的大尺寸和凝胶化行为以及低生物降解率可能导致基质中多糖的更高积累潜力[24, 25]。比如活性污泥中具有很好的团聚性和沉降性的菌胶团[26], 就是动胶菌属(*Zoogloea*)大量分泌胞外多糖而致。本试验虽然没能在开路和闭路系统的出水中检测到EPS, 但是从

孔隙率和过滤速率来看, 闭路系统的堵塞情况的确得到了一定程度的缓解, 其净化效果亦优于开路组。其实, EPS的分泌与降解是一个动态发展过程。电场对EPS的作用是微妙的, 可以降低EPS含量并改变其有机物组成, 避免由EPS过量积累引起的生物堵塞[14, 17]。

总的来说, 本试验采用嵌入电极的方式, 研究无植物CW系统生物堵塞原位缓解的结果还是较好的。随着电极材料的发展, MFC系统的产电性能将会大幅提高, 那么其在原位缓解人工湿地生物堵塞方面将具有较好的应用潜力。另外, 湿地植物对产电性能、污水处理性能均有影响, 植物根系的生长

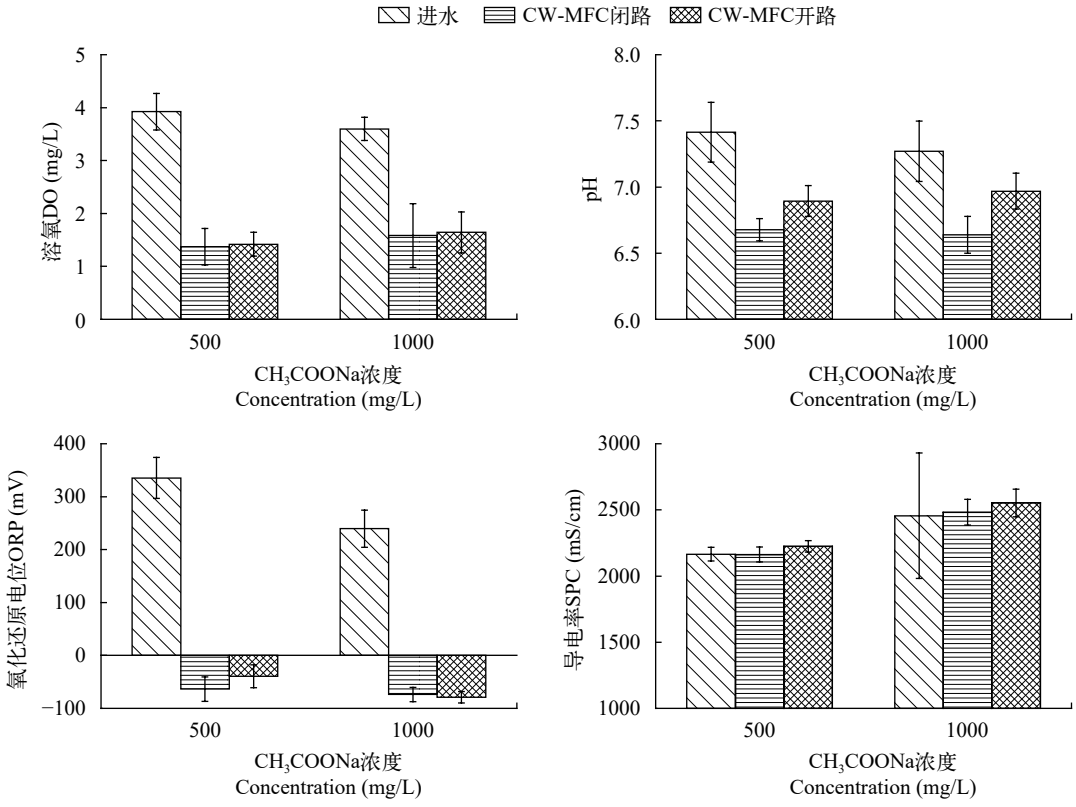

图 5 CW-MFC系统进出水pH、DO、SPC和ORP

Fig. 5 pH, DO, SPC and ORP of the inflow and outflow

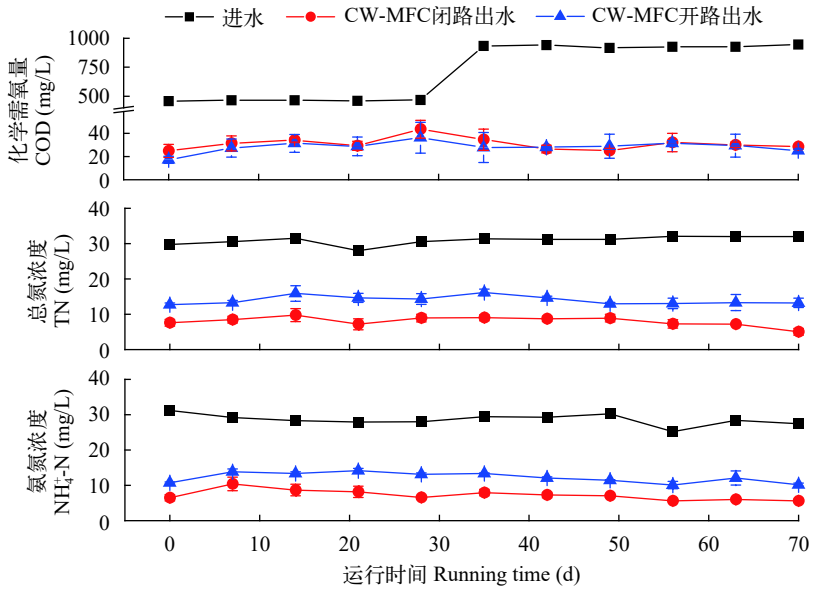

图 6 CW-MFC系统进出水COD、TN和NH_4^+-N浓度

Fig. 6 Concentrations of COD, TN and NH_4^+-N in influent and effluent from CW-MFC systems

和根系分泌物对基质堵塞也有重要影响, 在后续试验中还需深入研究。

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STUDY ON RELIEVING BIO-CLOGGING WITH THE HYBRID SYSTEM OF CONSTRUCTED WETLAND AND MICROBIAL FUEL CELL

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Abstract: Extracellular polymeric substances (EPSs) are biosynthetic polymers from prokaryotic (bacteria, archaea) and eukaryotic (algae, fungi) microorganisms, which either form (loose or tight) slimes around the microbial cells or are excreted as discrete gels to the surrounding environment. In biofilm systems, EPSs are mainly responsible for binding cells and other particulate materials together. A large secretion of EPS will continuously form a cohesive dense layer on the surface of the matrix. Therefore, the gradual accumulation of EPSs easily causes the severe bio-clogging of constructed wetlands (CWs) running for a long time. Microbial fuel cells (MFCs) could degrade the macromolecular organics EPS by the electrochemically active bacteria (EAB) colonizing on the surface of the electrodes and inhibit the large secretion of exopolysaccharides from microorganisms by the weak electric field formed during the electro-genesis process. Hence MFCs are supposed to be able to control the bio-clogging. In order to relieve the bio-clogging of CW system, this study constructed a CW-MFC hybrid system by embedding the anode and cathode electrodes into different depths of vertical direction of CWs. Two experiment groups with three parallels were set up in the closed circuit mode and the open circuit mode, respectively. The whole experiment consists of two stages. The first is bio-clogging simulation, and the second is bio-clogging mitigation. In the first stage, the excessive sludge from the sewage treatment plant was added to reactors to simulate the bio-clogging of constructed wetlands. When the obvious banked-up phenomenon happened and inconspicuous change of water filtration velocity were observed, the experiment system was regarded as clogging. In the second stage, the experiment lasted 70 days with two different influent concentrations. From the 1st to 35th day, the influent concentrations of TN, TP and COD were approximately 30, 2.5 and 500 mg/L, respectively. From the 36th day to the end, the TN and TP concentrations in the influent remained the same, while the COD concentration increased to 1000 mg/L. The porosity and the filtration velocity were used as the clogging index, the output voltage, the polarization curves and power density curves were used to evaluate the electro-genesis performance, and the removal rates of COD, TN and NH_4^+-N were used to assess the purification effect. The results showed that in CW-MFC circuit system, the change of filtration velocity (porosity) is bigger than that in open system. This gap was bigger in higher influent COD, which proves the relieved clogging in CW-MFC circuit system. The high removal rate of TN and NH_4^+-N indicated a good enrichment of denitrifies in circuit system. On the one hand, the higher the COD influent, the lower the COD effluent. On the other hand, the amount of output voltage or the converted electrical energy did not increase. This implied that more COD was utilized to sustain the growth of the heterotrophic denitrifies. In conclusion, the CW-MFC system can achieve a certain degree of the bio-clogging. Further research will have good application potential as in situ mode.

Key words: Constructed wetland; Bio-clogging; Porosity; Filtration velocity; Microbial fuel cell