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利用缓释碳源强化生物反硝化脱氮的研究进展

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摘要 水体硝酸盐污染对生态平衡和人体健康产生严重威胁, 利用微生物异养反硝化作用去除水体硝酸盐是目前的主流方法, 此过程通常需要投加额外碳源保证反硝化进行完全。传统商业碳源短板日渐显露, 缓释碳源作为一种新型固体碳源, 兼具经济性和高脱氮性能, 受到大量关注。从缓释碳源研究的必要性出发, 对其种类与反硝化效果、改性方法与成本分析、反应机制与微生物群落变化进行了全面详细的介绍, 总结了缓释碳源在协同处理其他污染物和工艺耦合方面的研究进展, 并对其未来研究方向提出建议, 以期缓释碳源促进反硝化脱氮的可行性探究提供全面的参考。

关键词 天然碳源 合成碳源 固相反硝化 微生物群落结构 协同处理 工艺耦合

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Advances in Research of Enhanced Biological Denitrification for Nitrogen Removal by Use of Slow-Release Carbon Sources

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Abstract Nitrate pollution in water bodies poses a serious threat to ecological balance and human health. The use of microbial heterotrophic denitrification to remove nitrate from water bodies is currently the mainstream method, which usually requires the addition of extra carbon sources to ensure complete denitrification. As the shortcomings of traditional commercial carbon sources become increasingly apparent, slow-release carbon sources, as a new type of solid carbon source, have received a lot of attention due to their economy and high denitrification performance. Starting from the necessity of slow-release carbon sources, a comprehensive and detailed introduction of their types and denitrification effects, modification methods and cost analysis, reaction mechanisms and microbial community changes is presented. The research progress of slow-release carbon sources in co-treatment with other pollutants and process coupling is summarized, and future research directions are suggested, with a view to providing a comprehensive reference for exploring the feasibility of promoting denitrification with slow-release carbon sources.

Keywords natural carbon source synthetic carbon source solid-phase denitrification microbial community structure co-treatment process coupling

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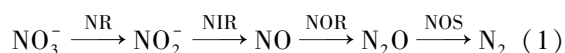
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硝酸盐污染是一类严重的环境问题, 它会影响到地表水和地下水的质量, 进而威胁人类和生态系统的健康。调查数据显示, 2016年我国河水中硝酸盐超标率达7.83%^[1]。根据我国《地表水环境质量标准》(GB 3838—2002), 地表水的硝酸盐质量浓度应低于45 mg/L, 根据《生活饮用水卫生标准》(GB

5749—2022),地下水的硝酸盐质量浓度应低于 89 mg/L^[2-3]。然而,近年来地表水和地下水受到的污染逐渐加剧,尤其是近十年来城市化、工业化的快速发展以及人口的增加,导致硝酸盐含量持续升高^[4]。沿海地区和内陆的地下水也遭受了不同程度的污染^[5-6]。氮超标排放不仅会导致富营养化^[7],还可能间接产生健康风险,如高铁血红蛋白血症、胃肠道癌、不良妊娠现象、甲状腺疾病以及神经管缺陷等^[1,8-10]。硝酸盐治理刻不容缓。

硝酸盐的去除方法有物化法(反渗透、电渗析、离子交换、化学脱硝等)和生物法[自养或异养反硝化、硝酸盐异化还原成铵(DNRA)等],其中,微生物异养反硝化是目前污水脱氮中最经济有效的方法^[11]。微生物异养反硝化作用,是反硝化菌利用硝酸盐还原酶(NR)、亚硝酸盐还原酶(NIR)、一氧化氮还原酶(NOR)、氧化亚氮还原酶(NOS)的催化作用将 NO₃⁻ 依次还原成 NO₂⁻、NO、N₂O,最终还原为 N₂ 排放,过程如式(1)^[12],该过程需要有充足的碳源为反硝化菌提供电子。常用的传统外加碳源包括乙酸钠、葡萄糖、甲醇等,这些碳源价格昂贵。污水处理厂在低碳氮比运行条件下,碳源的药剂成本在药剂总成本中的占比超过 70%^[13]。因此,亟需寻求一种兼具经济性和高脱氮性能的替代碳源。



缓释碳源作为一种来源广、价格低且能缓慢释放有机碳的新型固体碳源,在近年来的研究中颇受欢迎。利用天然纤维素类碳源(天然碳源)和合成可生物降解碳源(合成碳源)作为反硝化电子供体,可以强化系统的脱氮效果^[14-16]。本文总结了缓释碳源的种类及反硝化效果、作用机制及微生物群落结构、协同处理及工艺耦合研究,并对其未来的发展趋势进行展望,以期缓释碳源的脱氮效果研究提供参考。

1 缓释碳源种类和特点

缓释碳源按照其来源不同一般可以分为天然碳源、合成碳源以及混合碳源。

1.1 天然碳源

1.1.1 天然碳源的反硝化效果研究

天然碳源通常指天然木质纤维素材料,主要由纤维素(35%~55%)、木质素(15%~40%)和半纤维

素(5%~25%)组成^[17]。全球每年生产 10 亿~500 亿 t 干木质纤维素,约占全球生物质产量的 1/2^[18]。天然碳源可以作为反硝化的电子供体,稻草、秸秆、玉米芯、木屑、大米等都是近年来研究的热点^[19-22]。表 1 列举了不同种类的缓释碳源对硝酸盐的去除情况。天然碳源富含纤维素,在生物降解后可释放溶解性有机碳源,能够有效去除水中的硝态氮。序批式反应器(SBR)在添加玉米芯作为碳源后,总氮去除率从(45.46% ± 0.05%)提高到(70.32% ± 0.76%),脱氮优化效果显著^[20]。丝瓜络、麦秸、玉米芯、稻草具有较好的碳释放能力,作为外加碳源时也表现出良好的反硝化潜力(105.3~140.1 g N/g),膜生物反应器(MBR)在分别添加水稻秸秆、丝瓜络和玉米芯后的脱氮效果均得到有效强化,与对照组相比,总氮去除率分别提高 25.48%、19.53% 和 38.90%^[21],说明利用天然纤维素类物质替代传统反硝化碳源具有可行性。

1.1.2 改性天然碳源的反硝化效果研究

天然木质纤维素结构由木质素、纤维素和半纤维素镶嵌而成,而木质素的复杂性和难降解性增加了其降解难度^[23]。利用化学法(如酸处理、碱处理等^[24-25])对天然碳源进行改性,可以改善其有机碳释放能力^[26-27]。化学改性可以去除木质素和/或半纤维素,并在较小程度上降低纤维素组分的聚合度和结晶度^[23],提高纤维素的生物降解性。Li 等^[28]用 2% NaOH 溶液对丝瓜、玉米芯和秸秆进行改性后,其有机碳释放量翻倍,反硝化潜力从 112.04~157.94 mg/g 增加到 233.82~250.69 mg/g。不同植物对同种改性方法响应不同。美人蕉、芦竹、香茅、香蒲在进行相同碱处理后的表面粗糙度、释碳性能和反硝化性能均有所差异,香茅和芦竹的表面粗糙度和有机碳释放量最大,作为碳源的脱氮效果最佳,平均硝态氮去除率分别达到 89.46% 和 90.52%^[24]。碱处理能够明显减少天然纤维素碳源内的木质素含量^[29]。碱的使用可以促使酯和糖苷侧链降解,进而导致木质素结构改变、纤维素膨胀、纤维素部分消晶^[30-31]和半纤维素部分溶剂化^[25,32]。也有研究^[33]发现,改性后的天然碳源虽然有机碳释放量增大,但反硝化效果没有明显改善,且会释放 N₂O。因此,如何适当应用改性方法需要进一步研究。

1.1.3 天然碳源二次污染物的释放

天然碳源的成分复杂,有机碳、氮、磷和色度的

释放机制不清晰,其在降解或释放过程中可能会产生二次污染,对污水处理造成不利影响。多项研究^[34-36]表明,秸秆类碳源(如小麦秸秆、玉米秸秆、稻草秸秆等)在反硝化过程中普遍会有色度的产生,稻草秸秆的总氮释放量可达(20.77±1.67) mg/(g·d)^[34]。在实际应用中,外加碳源释放的过量总氮可能会对反硝化产生不利影响^[20],产生的色度对后续工艺也提出了额外要求^[35]。除此之外,缓释碳源的使用也可能会造成温室气体的排放。玉米芯、麦秸和绿色废弃物虽然具有较好的反硝化脱氮效果,但在反应过程中均有 N₂O 产生,其中玉米芯还会释放大量总有机碳(TOC)和 CH₄^[37]。因此,在实际生产中应用天然碳源作为反硝化碳源和生物膜载体时,除了要关注其有机碳释放特性和反硝化效果,也要充分考虑二次污染物的产生。天然碳源释放的有机物占总质量的主要为大分子有机物以及含双键有机物,其主要成分为色氨酸、黄腐酸及少量蛋白质,在后续的反硝化过程中被全部降解利用^[38]。而未浸出的组分以木质素及含木质素的纤维素为主,由于不含硫元素,天然木质素在消毒过程中不会产生有害副产物^[39]。

1.2 合成碳源

1.2.1 合成碳源的反硝化效果研究

合成碳源是指人工合成可生物降解聚合物,其特点是不溶于水,容易被微生物降解,可为反硝化恒定提供有效碳源^[40]。聚羟基链烷酸酯(PHA)、聚3-羟基丁酸酯(PHB)、聚己内酯(PCL)、聚丁二酸丁二醇酯(PBS)、3-羟基丁酸酯-共-3-羟基戊酸酯(PHBV)等常被用于固相反硝化研究^[41-49],且具有较好的脱氮效果。PHA和PHB是微生物自身的储存物质,容易被自然界存在的微生物代谢^[50],绿色塑料PHBV表面含有C-O、C=O等亲水基团,有利于微生物附着生长和降解利用,其作为电子供体和微生物载体的反应系统,反硝化启动快、有机物出水剩余浓度低、反硝化速率高、脱氮效果持续稳

定^[41,45,51]。PCL的反硝化效果虽不及PHBV^[47],但因经济优势也常被用作固相反硝化研究^[52]。PCL作为固相反硝化生物滤池的过滤介质和碳源处理污水处理厂二级出水,反硝化速率最大可以达到3.80 g N/(L·d),且在8℃的低温环境下反硝化速率依旧能保持在1.2 g N/(L·d),脱氮效果颇佳^[47]。

1.2.2 合成碳源的经济成本分析

脱氮工艺的成本主要由技术设备成本、维修成本和基质价格决定^[50]。与甲醇、乙酸钠等常用的传统碳源相比,PCL和PHB的药剂成本相对较高^[53],但对设备和装置的要求较少。在运行阶段,合成碳源的有机碳释放是随微生物对电子供体的需求降解产生,不需要对其进行额外的过程控制和水质连续监测操作,因此,虽然合成碳源的生产成本略高,但其在设备成本、运维操作上更为简单,且在运输和运行过程中基本不存在易燃性和毒性等安全风险^[47]。然而由于缺乏实际应用数据,无法准确地对使用合成碳源的脱氮工艺进行成本计算。未来随着可生物降解聚合物合成工艺的发展优化,合成碳源可能会逐渐实现量化生产,生产成本会日益降低,其经济性将会得到极大的提高,在反硝化处理中的应用将会愈发广泛^[40]。

1.3 混合碳源

为了在经济成本和脱氮效率方面取得平衡,研究人员将具有优越反硝化性能的合成碳源与经济有效的天然碳源共混,制备新型环保经济可生物降解复合物。由PHBV/竹粉混合物支撑的反硝化系统能够快速启动,硝酸盐去除率高,过程中几乎无亚硝态氮积累,并且混合物的使用可以降低基质成本,是一种经济且高效的碳源选择^[54]。Yang等^[55]的研究发现聚乳酸(PLA)/淀粉共混物在质量比为1:1时反硝化速率最大为4.608 g N/(L·d),出水TOC的累积较少,具有良好的环境适应性和脱氮性能。混合碳源能够结合天然碳源和合成碳源的优点,降低二者的不良影响,具有广阔的发展前景。

表1 各类缓释碳源反硝化系统的反硝化性能

Tab. 1 Performance of All Kinds of Slow-Release Carbon Sources Denitrification Systems

碳源	进水水质	进水硝态氮质量浓度/(mg·L ⁻¹)	反应工艺类型	硝态氮去除率	反硝化速率	水力停留时间(HRT)/h
陈米 ^[22]	人工快渗系统出水	9.48~26.00	有机玻璃柱	33.43%~93.37%	-	2

(续表1)

碳源	进水水质	进水硝态氮质量浓度/(mg·L ⁻¹)	反应工艺类型	硝态氮去除率	反硝化速率	水力停留时间(HRT)/h
木屑 ^[19]	合成废水	50~100	人工湿地	63.6%~96.1%	-	48
木片 ^[56]	合成废水	50	两级上流塔反应器	75%	0.0189 g N/(L·d)	24
花生壳 ^[57]	合成废水	12.0~15.7	摇瓶试验	76.48%±4.06%	-	
瓦楞纸板 ^[58]	RAS 出水	-	SPD 反应器	82.88%	0.00074 g N/(L·d)	
稻壳 ^[59]	合成废水	50	生物电化学反应器	-	0.00034 g N/(L·d)	
木屑 ^[16]	合成废水	50	上流式 SPD 反应器	92.5%~96.4%	-	
玉米芯 ^[20]	合成废水	-	SBR	70.32%±0.76%	-	
木屑 ^[19]	合成废水	50~100	人工湿地	63.6%~96.1%	-	48
PHBV ^[41]	合成废水	15	有机玻璃填充柱	≥95%	0.77 g N/(L·d)	0.5~1.0
PBS ^[48]	RAS 出水	50~150	-	≥90%	(0.53±0.19) g N/(L·d)	
PCL ^[42]	合成废水	48~50	-	>95%	-	
PCL ^[43]	合成废水	25~50	-	>98%	-	
PCL ^[47]	二级出水	10.8~14.7	生物滤池	87.5%~99.6%	0.41~2.65 g N/(L·d)	
PHBV ^[45]	合成废水	7.8~25.2	生物滤池	90%~100%	1.003 g N/(L·d)	0.5
PBS ^[44]	合成废水	-	SBR	56.4% (总氮)	-	
PCL ^[46]	RAS 出水	89.3~101.68	上流式填充反应器	-	0.03467~0.1557 g N/(L·d)	6
PHBV/锯末 ^[60]	二级出水	7.65~12.46	上流式 SPD 反应器	91.65%±4.12% (总氮)	-	1.5
淀粉/PVA-SA ^[61]	合成废水	35.0~40.0	上流式连续填料床反应器	>94.3%	0.36~0.93 mg N/(g·d)	4
PHBV/PLA/稻壳 ^[62]	合成废水	30	填料床生物反应器	77%~98%	0.118~0.288 g N/(L·d)	2~6
PHBV/PLA ^[63]	合成废水	15.5	连续填充床反应器	94.11%	1.68 mg N/(g·d)	1
PHBV/PLA ^[64]	农田沟渠出水	5.05±0.26 (总氮)	水平地下流	64.5%±4.11% (总氮)	-	36
PCL/花生壳/PVA-SA ^[65]	合成废水	20	摇瓶试验	99.92%	0.0105 g N/(L·d)	120
PCL/淀粉 ^[66]	SBR 出水	19~22	垂直折流板 SPD 反应器	(33.1%±2.2%)~(89.1%±5.1%)	0.08~0.33 g N/(L·d)	2

注: * RAS 为循环养殖系统;SPD 为固相反硝化;PVA-SA 为聚乙烯醇海藻酸钠。

2 缓释碳源促进反硝化脱氮机制

2.1 有机碳底物转化机制

缓释碳源不溶于水,能够通过胞外水解酶降解产生可溶性有机单体或低聚物,以供反硝化作用利用,具体过程如图 1 所示。天然碳源水解可生成葡萄糖、阿拉伯半乳糖、木糖等低分子有机化合物^[67],乙酸、丙酸和丁酸等短链脂肪酸则是合成碳源降解后产生的主要有效碳源^[41,65,68]。大部分降解生成

的底物可以作为电子供体,为反硝化过程所用,改善污水中硝态氮的去除效果,也有部分碳源被用于厌氧消化产甲烷^[37]或好氧生物降解^[69]。

2.2 硝酸盐转化机制

在缓释碳源支持的固相脱氮体系中,存在多种硝酸盐转化机制。异养反硝化过程由不同生物酶催化完成,在固相脱氮系统中占据绝对优势。NO₃⁻经由膜结合硝酸还原酶(NAR)或周质硝酸还原酶

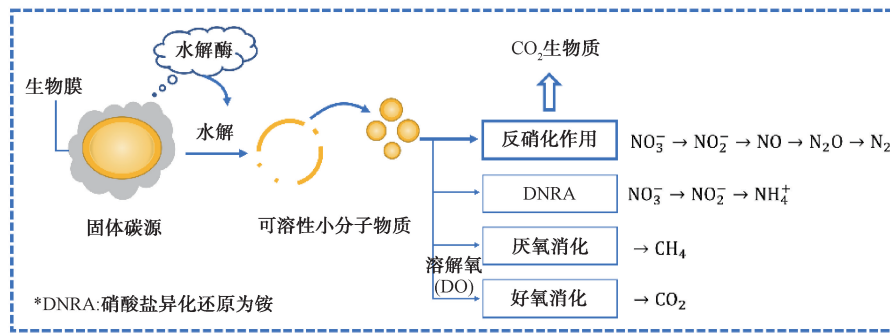


图 1 缓释碳源固相反硝化^[53]

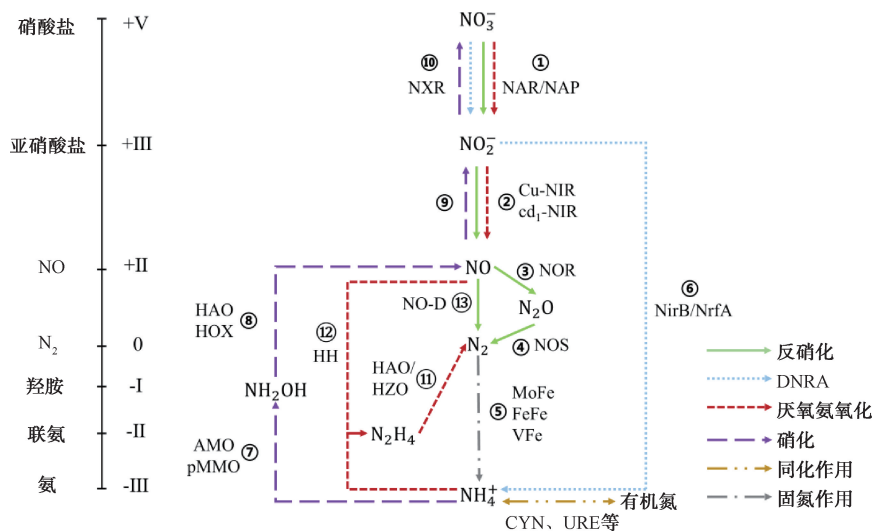
Fig. 1 Solid-Phase Denitrification by Slow-Release Carbon Sources^[53]

(NAP)催化还原成 NO_2^- 。含血红素的 cd_1 亚硝酸盐还原酶 (cd₁-NIR) 或含铜亚硝酸盐还原酶 (Cu-NIR) 进一步将 NO_2^- 还原成 NO , 这两种酶分别由 nirS 和 nirK 基因编码^[70]。NOR 和 NOS 依次将 NO 还原为 N_2O , 最终生成 N_2 排放, 整个过程如图 2 (①~④) 所示。相比其他氮氧化物还原酶, NOS 对溶解氧、pH 和硫化物等环境因素更敏感^[70], 在高硝酸盐、低有效碳环境中, 其电子消耗速率在所有反硝化酶中最低^[71], 容易导致不完全反硝化, 造成 N_2O 的积累与释放。DNRA [图 2 (①、⑥)] 过程是固相反硝化系统中出现的次要反应, 在 NO_3^- 还原为 NO_2^- 后经 NirB 或 NrfA 两种 NIR 异化还原为铵^[72], 其在

固相脱氮系统中贡献率低于 10%^[73]。亦有研究^[74] 发现, 固相脱氮系统中存在厌氧氨氧化菌, 但是否存在厌氧氨氧化过程的参与还有待验证。与常规反硝化相比, 缓释碳源可以为微生物附着场所, 形成生物膜, 增加微生物的种群多样性以及提高微生物浓度, 从而促进反硝化效果的提高^[75]。虽然微生物脱氮的原理是相同的, 但是添加不同的碳源, 其优势微生物存在一定的差别。具体情况在第 3 小节进行讨论。

3 缓释碳源反硝化系统中微生物群落变化

缓释碳源是一种替代传统碳源的新型固体碳源, 通过微生物的降解释放可溶性有机碳, 为反硝化细菌提供电子。



注: NXR 为亚硝酸盐氧化还原酶; NO-D 为一氧化氮歧化酶; HH 为胍水解酶; HZO 为胍氧化酶; HAO 为羟胺氧化还原酶; HOX 为羟胺氧化酶; AMO 为氨单加氧酶; pMMO 为微粒甲烷单加氧酶; MoFe 为钼铁固氮酶; FeFe 为铁铁固氮酶; VFe 为钒铁固氮酶; CYN 为氰酶; URE 为脲酶。

图 2 含氮化合物的微生物转换^[70,76]

Fig. 2 Microbial Conversion of Nitrogen-Containing Compounds^[70,76]

3.1 反硝化菌群变化

缓释碳源的加入会改变微生物群落的丰富度,但大部分系统中变形菌门依旧占据主导地位,且呈现增加趋势。在传统的污水处理系统中,微生物的优势菌门一般为变形菌门(Proteobacteria)、拟杆菌门(Bacteroidetes)、放线菌门(Actinobacteria)和绿弯菌门(Chloroflexi),其中变形菌是几乎所有反硝化研究中的优势菌门^[77]。表2列举了不同固相反硝化体系中的微生物组成情况,缓释碳源的加入为微生物提供了独特的附着生存环境,导致系统微生物群落结构发生变化,不同碳源主导的系统中菌群类型差异大,但优势菌门依旧是具有反硝化作用的变形菌。PCL作为碳

源应用于生物滤池三级脱氮处理后,其表面生物膜中微生物群落发生演替,微生物多样性有所下降,初始接种污泥中的 *Thauera*、*Nitrospira*、*Arenimonas*、*Dechloromonas* 等在碳源生物膜中几乎消失殆尽,而具有反硝化效果的变形菌相对丰度变大^[47]。在以玉米芯为碳源的反硝化体系中,拟杆菌、变形杆菌和厚壁菌是系统的主要菌门,占每个样本总丰度的 50.60%~89.48%,且随反应的进行,系统中的微生物结构呈现动态变化,隶属于拟杆菌门的 *Prevotella*_9 在反应结束时几乎消失,相对丰度从 49.31%降为 0.33%,而隶属于变形菌门的 *Dechloromonas*、*Denitratisoma* 和 *Bdellovibrio* 则普遍呈现增加的趋势^[20]。

表2 固相反硝化系统中主要的反硝化细菌

Tab. 2 Main Denitrifying Bacteria in Solid-Phase Denitrification System

碳源种类	反硝化细菌(属水平)	备注
PCL ^[47]	<i>Acidovorax</i> 、 <i>Bdellovibrio</i> 、 <i>Thermomonas</i> 、 <i>Leptothrix</i> 、 <i>Thauera</i> 、 <i>Simplicispira</i> 、 <i>Flavobacterium</i>	变形菌门和拟杆菌门为主要优势菌门
PCL ^[78]	<i>Comamonas</i> 、 <i>Betaproteobacteria</i> _Unclassified、 <i>Acidovorax</i> 、 <i>Gracilibacteria</i> _Unclassified、 <i>Stenotrophomonas</i> 、 <i>Alicyclophilus</i>	变形菌门和拟杆菌门为优势菌门
PBS ^[79]	<i>Simplicispira</i> 、 <i>Dechloromonas</i> 、 <i>Diaphorobacter</i> 、 <i>Desulfovibrio</i> 、 <i>Thauera</i> 、 <i>Pseudomonas</i> 、 <i>Denitromonas</i>	变形菌门为优势菌门,且随反应进行占比持续增加
PHBV ^[74]	<i>Dechloromonas</i> 、 <i>Desulfovibrio</i> 、 <i>Magnetospirillum</i> 、 <i>Azoarcus</i> 、 <i>Geobacter</i> 、 <i>Thaurea</i> 、 <i>Azonexus</i> 、 <i>Candidatus</i> <i>Accumulibacter</i>	变形菌门为优势菌门,其次为拟杆菌门
PHBV/木屑 ^[74]	<i>Dechloromonas</i> 、 <i>Italea</i> 、 <i>Bradyrhizobium</i> 、 <i>Ignavibacterium</i> 、 <i>Gemmatimonas</i> 、 <i>Candidatus</i> <i>Accumulibacter</i> 、 <i>Conexibacter</i> 、 <i>Nitrospira</i>	变形菌门为优势菌,其次为放线菌门;存在少量的厌氧氨氧化菌
玉米芯 ^[20]	<i>Prevotella</i> _9、 <i>Microbacter</i> 、 <i>Prevotella</i> _1、 <i>Genera</i> <i>Dechloromonas</i> 、 <i>Denitratisoma</i> 、 <i>Bdellovibrio</i>	微生物群落结构呈动态变化,以变形菌门为主的反硝化菌群不断增加
木片 ^[16]	<i>Flavobacterium</i> 、 <i>Cloacibacterium</i> 、 <i>Bacteroidetes</i> _Vadinha17_Norank	随着反应的进行,变形菌门取代拟杆菌门成为优势门

3.2 降解类微生物菌群变化

具备可以降解可生物降解聚酯的细菌是缓释碳源降解的基础。绿弯菌门可以将溶解性微生物产物(SMP)降解成碳水化合物和细胞材料,破坏蛋白质、多糖、纤维素等一些大分子物质^[79],在以PBS作为碳源的反应系统中比例有所增加^[80]。丛枝孢科亦被认为具有降解可生物降解聚酯能力,在PCL固相反硝化系统中占据优势地位,相对丰度为79.1%~73.3%^[47]。隶属于根瘤菌目的 *Devosia*、*Agrobacterium*、*Polymorphum* 和 *Bradyrhizobium* 被认为具有发酵纤维素、半纤维素等复杂有机物的能力, *Acidovorax*、*Simplicispira* 和 *Dechloromonas* 可以直接降解木质纤维素,经常在固相脱氮系统中被检测

到^[29]。这些微生物的存在可以促进缓释碳源的分解,从而为反硝化过程提供充足的有效碳源。

3.3 微生物群落随环境因素变化

微生物群落结构随环境条件(温度、溶解氧、pH、底物可利用性等)变化而不同。一般情况下,微生物对于系统中pH的变化会进行应激反应,可以在一定程度上维持pH的稳定性,且在污水处理过程中,一般会人为调控,使pH维持在一个合理范围内,所以pH对于微生物种群的影响相对较小。而温度、溶解氧、反应底物常常会随着季节、工艺、运行成本控制等因素影响,发生比较大的变化。当温度从30℃下降至13℃时,以PCL为碳源的中试反应器硝态氮去除率降低37%,不同温度下系统富集的

微生物存有较大的差异,低温系统富集了更多的 *Desulfomicrobium*、*Desulfoicrobium* 和 *Meganema*,而高温条件下富集有 *Simplicispira*、*Aquabacium* 和 *Acidovorax*^[81]。溶解氧对于微生物的种群影响是最明显的。文献^[82]表明,好氧条件下(DO 质量浓度大于 0.5 mg/L),促进碳源分解的主要是绿弯菌门(*chloroflexi*),相应的反硝化细菌为变形菌门,具体菌属包括 *Methyloversatilis* (16.6%)、*Chlorobium* (15.09%)、*Rhodopseudomonas* (8.17%);而厌氧条件下(DO 质量浓度小于 0.2 mg/L),厚壁菌门(*Firmicutes*)和放线菌门占据绝对优势。此外,具有水解酸化功能和反硝化功能的菌属 *Hydrogenophaga*、*Fimbriimonas*、*Pseudoxanthomonas*、*Acidovorax*、*Dysgonomonas* 通常相伴出现^[38,53,66,83]。Yang 等^[74]分别探究了以 PHBV、PHBV/木屑混合物为碳源的反硝化系统的脱氮性能差异和微生物群落结构差异,结果发现两系统中变形菌门均占主导地位,其余菌门的比例则存在明显差异,拟杆菌门是 PHBV 系统中的第二大优势菌门,而 PHBV/木屑混合物系统中的第二大优势菌门则为放线菌门,并且 PHBV/木屑混合物系统中还发现了广古菌门,可能与混合物的降解有关。文献^[84]表明,以 PCL 为缓释碳源时,优势菌属为湖沼菌属(*Limnohabitans* sp., 57.8%)和单胞菌属(*Simplicispira* sp., 为 9.5%),而以 PHA 为载体时,优势菌属为山冈单胞菌属(*Collimonas* sp., 24.4%)、湖沼菌属(*Limnohabitans* sp., 13.2%)和叶黄素单胞菌属(*Luteimonas* sp., 11.7%)。然而,造成这一现象的原因,还有待进一步探究。

在碳源表面形成的生物膜中,变形菌门作为常规的反硝化菌群依旧占据主导地位,并且在系统中呈动态增加,与缓释碳源降解相关的绿弯菌、食酸菌、脱氯单胞菌等细菌也广泛存在于固相反硝化系统中。微生物群落结构的变化为固相反硝化强化脱氮奠定了基础。

4 缓释碳源的其他研究与应用

4.1 固相反硝化与其他工艺耦合

在缓释碳源相关研究中,通常将其应用于人工湿地、生物滤池、上流式反应器等常规反应器,近几年,有研究者尝试将缓释碳源与微电解、厌氧氨氧化等工艺相结合,观察系统的反硝化脱氮强化效果。

研究发现固相反硝化碳源与铁碳微电解耦合系统能够释放充足的有效碳源^[85],系统内部同时存在自养和异养反硝化菌群,在最佳运行条件下,系统对总氮和总磷的去除率能够达到 94.72%、99.10%^[86],污染物去除效果得到明显改善。Zhao 等^[87]将微生物燃料电池(MFC)和固相反硝化系统(DBR)耦合构建的新型电化学系统,对甲基橙、Cr(VI)、硝态氮的平均去除率分别提高 39.9%、26.5%、22.8%。除此之外,缓释碳源应用于厌氧氨氧化系统对总无机氮的去除率能够高达 98.5%,在处理富氮废水方面具有良好的应用前景^[88]。综上,通过固相反硝化与其他工艺的耦合可以有效强化系统的脱氮效果。

4.2 协同去除硝酸盐和其他污染物

通常情况下,废水中除常规的污染物外,还可能会存在农药、药物与个人护理用品(PPCPs)、难降解有机化合物等其他物质的污染。研究表明,缓释碳源在协同去除这些特殊污染物方面也能发挥效用。在已有的探究中,缓释碳源对于阿特拉津的去除以吸附为主,在吸附量到达对应缓释碳源的吸附上限之后,多余的阿特拉津甚至会对微生物的活性造成影响^[89];对氯苯酚、四氯化碳、难降解有机物中的硝酸盐的去除以微生物为主,微生物去除占比接近 100%^[90-92];对氟乐灵、灭硝硫磷、硫丹的去除属于微生物转化与缓释碳源吸附的协同作用,其中,微生物的转化作用分别占 96%、87%、68%^[93]。目前普遍认为缓释碳源协同处理其他污染物的机理是碳源吸附和生物降解的共同作用,而究竟是哪种作用占主导地位仍存在争议。

5 结论与展望

缓释碳源作为一种新型固体碳源,在促进反硝化脱氮方面具有较好的发展前景。

(1)天然碳源来源广泛、价格经济,能够提高系统的脱氮性能,但存在二次污染物释放的风险,使用时需充分衡量其脱氮效果及可能造成的二次污染。

(2)合成碳源普遍具有较好的反硝化效果,但成本要明显高于天然碳源。

(3)缓释碳源经由微生物降解后释放小分子量有机物,为反硝化过程提供电子。

(4)变形菌门是传统反硝化系统中的优势菌门,在缓释碳源支撑的反硝化系统中依旧占据主导地位,绿弯菌、食酸菌、脱氯单胞菌等的存在为缓释

碳源的降解提供了可能。

(5) 缓释碳源不仅可以促进反硝化脱氮, 还可以通过吸附和生物降解协同去除农药、PPCPs 等特殊污染物。固相反硝化与电化学、厌氧氨氧化的耦合以其优秀的脱氮性能表现出良好的应用前景。

然而由于缓释碳源结构和成分复杂, 其在生物降解和反硝化过程中的反应机理需要更加深入地探索和研究, 且目前对于缓释碳源的研究主要集中在实验室研究水平, 缺少工程试验数据支撑, 亟待不同层面的研究。未来对于缓释碳源研究方向提出以下展望。

(1) 从反应动力学、微生物群落结构以及基因层面深入全面探究缓释碳源反硝化脱氮机理。

(2) 探究缓释碳源的二次污染释放情况, 包括出水中的二次污染及温室气体的排放情况等, 对缓释碳源的可利用性做更加全面的评估。

(3) 增加工程试验, 为缓释碳源的工程应用提供功能更多的数据和案例支撑。

(4) 利用生物信息学与机器学习开发, 面向特定环境与功能微生物的缓释碳源设计合成方法。

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