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功能基质强化人工湿地控制农业氮素面源污染研究进展

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摘要: 农业氮素污染所致的面源污染已引发黑臭水体等水环境问题，严重阻碍了农村生态文明建设。本文围绕生物炭、铁元素和微生物燃料电池等功能基质在人工湿地中的应用，系统综述了强化人工湿地系统的深层机制及研究进展。生物炭和铁元素能够强化微生物之间以及微生物与植物之间的联系。微生物燃料电池对微生物之间协作的强化作用更为显著。本综述聚焦氮素导致的农业面源污染现状，提出通过功能基质强化人工湿地实现有效控制的策略，为功能基质在人工湿地中的实际应用及农业氮素面源污染的有效控制提供建设性建议，以期为未来农村生态文明建设提供重要的理论支持。

关 键 词: 氮素面源污染；新型人工湿地；功能基质；黑臭水体

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农业面源氮素污染不仅使水体恶化，而且会造成地下水硝酸盐含量超标，导致水体富营养化[1]。研究表明，80% 的水体和 50% 的全球陆地面积受到农业面源污染的影响[2-3]。面源污染能够形成黑臭水体，严重影响农村地表水和地下水环境，已经成为农村生态文明建设的亟待解决的水环境问题[4]。氮素造成的面源污染已经成为当今世界水质恶化最主要的原因[5]。为了确保农业可持续发展，面源污染正在从源头控制到末端治理被控制[6]。

人工湿地 (CW) 被认为是一种有效的末端治理技术，通过模拟天然湿地设计，由大型植物、基质和微生物组成，主要分为自由水面人工湿地 (FWSCW)、潜流人工湿地 (SFCW) 和垂直流人工湿地 (VFCW)[7-8]。相对于污水处理厂建设，人工湿地在农村地区建设相对容易且接受程度高，能够有效控制农村氮素面源污染，具有耐用性、低外部能源需求、易于操作和维护的特点[9]。基质是人工湿地的重要组成部分，以多种角色参与系统氮素去除、转化去除污染物、促进植物生长和确保生物膜黏附[10]。部分基质能够通过提供电子转移直接参与微生物活动[11-12]。但是，人工湿地的基质选择存在盲目性，经常利用以往经验和主观判断直接使用基质，并且基质使用较为传统和单一，未能建立植物–填料–微生物之间有效联系，导致系统中氮的去除效果不理想[13]。这限制了人工湿地在氮素污染控制中的有效性。

近年来，功能基质在人工湿地中的应用显著提升了其氮素去除能力，通过有效强化各组成部分之间的相互联系，获得了研究人员的广泛关注。本文从提高氮素污染物去除效率的角度出发，系统综述了功能基质强化人工湿地的机制及研究进展，旨在为农业氮素面源污染控制提供有效的技术手段，并为人工湿地在农业氮素面源污染控制中的实际应用提供重要的理论支持。

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1　功能基质强化人工湿地的研究进展

传统人工湿地基质研究多关注于吸附和转化污染物的效果，植物、微生物和基质之间相对独立[14]。为了提升人工湿地氮去除效果，强化各个部分之间协作的功能基质应运而生[15]。对于功能基质强化人工湿地氮去除方面，研究最广泛的为生物炭[16-17]、铁元素[18]以及微生物燃料电池(MFC)[19]。生物炭能够作为植物和微生物之间有效的媒介，促进污染物的传递与转化[8-9]。铁元素由于其在地球上的丰富性和广泛的化合态，常用于强化人工湿地的研究[10]。微生物燃料电池与人工湿地结合，通过电化学途径促进系统氮去除过程同样具有显著效果[11]。这些功能基质将人工湿地中植物、微生物和污染物之间紧密联系，在人工湿地氮素污染控制中显示出巨大潜力。

1.1　生物炭基人工湿地

生物炭是在限氧高温环境制备多孔材料，前驱体 (指用于制备生物炭的原始生物质材料) 多为农业废弃物，生物炭在人工湿地中的作用机理 (图 1) 主要包括静电吸引、沉淀、分子间 π-π 骨架、离子交换和静电吸引等[20]。在生物炭强化系统微生物方面，研究表明，生物炭在低温下增加与反硝化过程相关功能基因，如 *nirS*、*nirK*、*napA*、*nxrA* 和 *narG*，强化人工湿地中总氮 (TN) 的去除[21]。厌氧氨氧化 (Anammox) 作为新型脱氮途径，人工湿地与 Anammox 组合能够有效降低污水中的 TN，生物炭已被发现能够强化 Anammox 功能菌并促进相关微生物生长[22-23]。Ajibade 等[24] 在以生物炭作为基质的人工湿地系统中，通过富集 *amx* 基因 (厌氧氨氧化标志物) 提升 Anammox 菌群丰度 (高达 14%)，使 TN 去除效率达到 90%。改性生物炭功能性更加显著。Jia 等[25]利用 $FeCl_3·6H_2O$ 对竹炭进行改性，发现改性生物炭能够加速胞外电子转移，参与微生物代谢活动。另外，生物炭能够帮助人工湿地植物完成吸收，强化植物根系分泌物产生，为根际微生物代谢提供营养物质，进一步提高污染物去除率[26-27]。

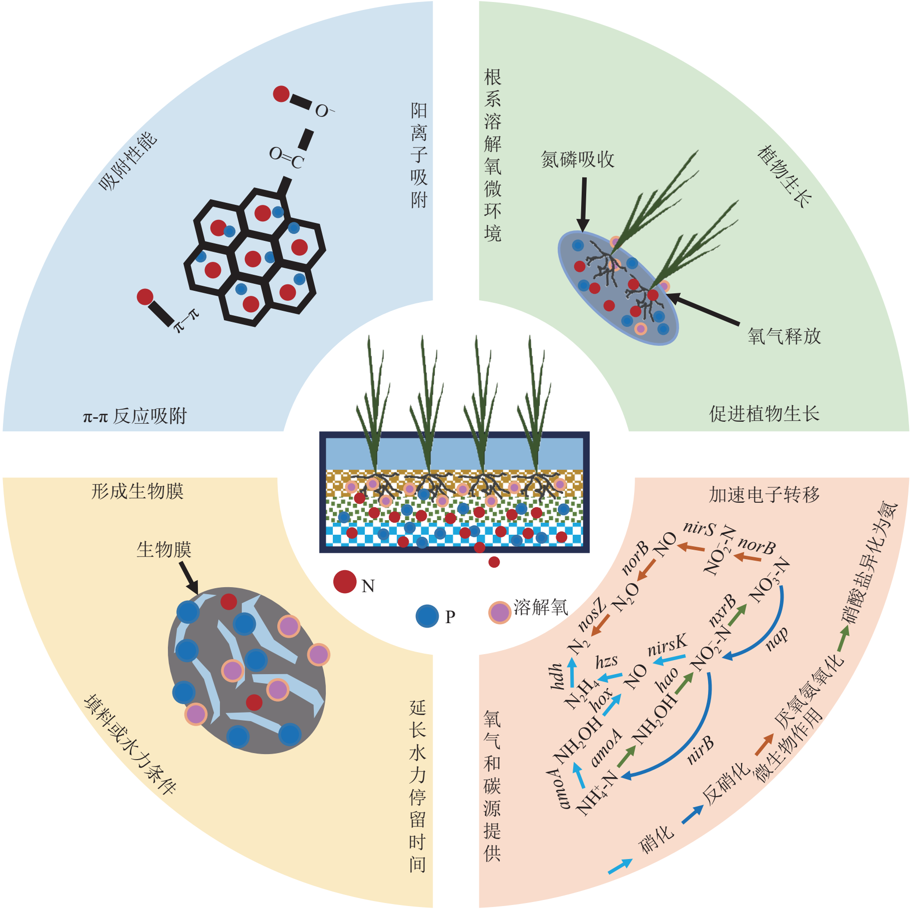

图 1　生物炭强化人工湿地氮去除机制[17]

Fig.1　Mechanism of nitrogen removal in constructed wetlands enhanced by biochar[17]

生物炭强化人工湿地氮素去除已经有较多研究 (表 1)[28]。生物炭在人工湿地中可以与砾石、沙子或沸石混合作为基质，也可以分层填充[29]。研究发现，相对于传统基质填充，生物炭与砾石以 1∶4 的体积比混合，对 TN 的去除率高达 91%[30]。另外，相对于仅用沸石的人工湿地，将生物炭与沸石混合可以使 TN 去除率提高 63%[29]。但是，生物炭的添加量会显著影响其强化效果。当生物炭体积填充比较低 (2%) 时，人工湿地对 TN 的去除率仅为 12%[31]。而将 10% 生物炭与砾石分层填充时，能够有效缓解人工湿地堵塞问题[25]。生物炭受制于前驱体影响，并不是所有类型的生物炭都能提升人工湿地的效能。Meng 等[28] 发现，与对照组相比，在 VFCW 中添加污泥生物炭，TN 去除效率仅为 28%(在 0.52 g/m³·d 的进水负荷条件下)。另外，Feng 等[31] 研究也发现类似现象，与对照组 (41%)相比，生物炭改良的 CW 中 TN 的去除率仅为 34%。其原因主要是难降解有机物 (如腐殖类组分) 造成的碳源不足，以及生物炭与硝态氮 (NO_3^--N) 之间的排斥。因此，生物炭受多种制备因素的影响，如何减少生物炭制备成本及合理化设计是其在人工湿地中实际应用重要研究方向。

表 1　生物炭在多种类型人工湿地中污染物去除性能

Table 1　Pollutant removal performance of biochar in various types of constructed wetlands

序号	人工湿地类型	废水类型	前驱体类型	水力停留时间/d	植物	去除效率/%				参考文献
						氨氮	总氮	总磷	化学需氧量	
1	垂直流人工湿地	低碳氮比	竹子(Bambusoideae)	3	水芹(*Oenanthe javanica*)	99.10	52.70	–	94.90	[32]
2	垂直流人工湿地	养猪废水	竹子	3	黄菖蒲(*Iris pseudacorus*)	88.62	31.37	–	67.76	[31]
3	混合人工湿地	模拟污水	竹子	3	芦苇(*Phragmites australis*)	98.80	95.70	100.00	89.10	[33]
4	潜流人工湿地	模拟污水	桤木(*Alnus glutinosa*)	2	香蒲(*Typha orientalis*)	–	20.00	22.50	75.00	[34]
5	垂直流人工湿地	真实污水	污泥	2	互花米草(*Spartina alterniflora*)	95.90	28.00	59.50	83.80	[28]
6	潜流人工湿地	模拟污水	芦竹(*Arundo donax*)	7	水芹	93.95	85.62	–	44.16	[35]

注：–表示相应参考文献中未检测该指标。

1.2　铁强化型人工湿地

铁在人工湿地系统中主要通过参与新型自养微生物脱氮途径和促进植物生长，实现强化系统污染物去除[11]。新型自养反硝化细菌可以利用铁作为电子供体，在厌氧或缺氧环境中将 NO_3^--N 和亚硝态氮 (NO_2^--N) 还原成氮气 (N_2)，即硝酸盐依赖的铁氧化过程 (NDFO)，相对于传统反硝化过程，NDFO 能够显著减少碳源使用和温室气体产生[36]。另外，厌氧铁氨氧化 (Feammox) 是首次在湿地生态系统中发现的新型自养脱氮途径，通过将 Fe^{3+}和氨氧化物还原为 N_2、NO_2^--N 或 NO_3^--N，缩短了氮循环过程路径，从而提供了一种新的脱氮途径[37]。事实上，由于铁的多种化合价态，氧化还原过程中能够为微生物提供生长所需能量，从而直接参与生物脱氮过程。传统反硝化过程利用有机碳源作为电子供体，将 NO_3^--N 转化为 N_2 (图 2)。对于植物影响方面，Fe 是植物生长和发育的必需元素，并且在诸如呼吸、光合作用、氮同化和固定、激素合成、DNA 合成以及活性氧的形成过程中起重要作用[38]。Fe 的氧化还原过程参与各

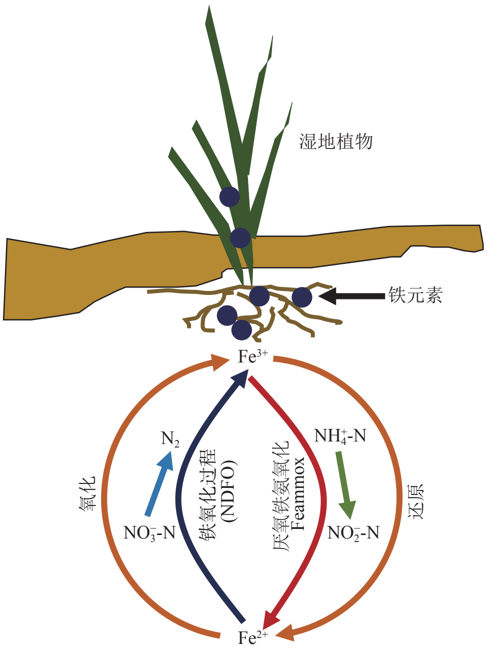

图 2　人工湿地中铁元素参与氮转化途径[18]

Fig.2　Pathways of nitrogen transformation involving iron in constructed wetlands[18]

种生理反应，Fe^{2+}对植物和微生物生长有显著影响，最佳剂量 Fe^{2+}可作为植物中氨基酸和酶活性的调节剂，从而优化微生物群落的丰度和分布以去除污染物[39]。添加铁元素的人工湿地系统中，许多植物在根表面能够形成铁膜，以适应淹水环境，显著强化厌氧环境中污染物去除过程[38]。 同时铁元素能够通过与 Fe^{3+}和 Fe^{2+}的相互作用除去磷酸盐，形成农业生产所需的磷肥，如 $FePO_4$ 和 $Fe_3(PO_4)_2$[38]。从该角度看，铁元素的引入在一定程度上具有农业生产效益。

利用铁进一步优化湿地系统的净化效果在经济上是可行的。研究发现，向湿地系统中添加铁屑可以增强硝化、反硝化作用和磷的去除，约 95% 的有机物、94% 的总磷 (TP) 以及超过 70% 的 TN 被去除 (表 2)。但是在人工湿地系统中，碳源往往不足，导致反硝化过程不完全，而不完全反硝化过程是上述人工湿地系统氧化亚氮 (N_2O) 产生的主要过程。在人工湿地中投加 Fe^{3+}可以通过 Feammox 进一步去除湿地系统中的氨氮 (NH_4^+-N)，Fe^{3+}还原产生的 Fe^{2+}可以继续作为电子供体促进系统的反硝化过程，有效降低水体中 NO_3^--N 的积累。同样地，Fe^{2+}浓度也能够影响湿地系统对 TN、NO_3^--N 和 NH_4^+-N 等污染物去除。以 Fe^{2+}为电子供体的反硝化工艺可以减少有机碳源投加，减少 NO_2^--N 和过量有机物积累，有效地去除硝酸盐[38]。但是，以上研究未能深入探索铁元素氮去除过程机制，而仅停留在氮去除效能提升层面。Cheng 等[41] 将 NDFO 和 Feammox 结合到同一人工湿地系统中，实现了对富营养化水体的有效净化，TN 的去除效率达 60%。但是其研究不仅涉及到铁元素，其中反应过程还涉及生物炭添加，因此无法将 TN 去除提升仅归于 Fe 元素存在。另外，其研究未对 NDFO 和 Feammox 过程的温室气体排放量进行探索，但这 2 种途径确实提高了人工湿地的氮去除效能。以上研究多关注于新脱氮途径的发现，但是铁对系统中植物和功能微生物具体影响机制还有待深入探讨。最重要的是，水体中铁化合物过多会严重影响水质，并且人体过多摄入严重时会引起铁中毒。因此，对于铁元素引入人工湿地，除了考虑其作用机制外，还应考虑其安全性。

表 2 铁元素强化人工湿地污染物去除性能

Table 2 Iron element enhanced constructed wetlands pollutant removal performance

序号	人工湿地类型	废水类型	铁材料	水力停留时间	植物	去除效率/%				参考文献
						氨氮	总氮	总磷	化学需氧量	
1	潜流人工湿地	生活污水	ZVI	3 d	鸢尾花(*Iris germanica*)	66.68	71.98	93.54	95.82	[40]
2	垂直流人工湿地	模拟污水	Fe^{2+}	2 d	美人蕉(*Canna indica*)	−	66.73	−	86.13	[42]
3	潜流人工湿地	模拟污水	Fe^{2+}	9 d	芦苇	−	81.28	−	84.00	[43]
4	垂直流人工湿地	模拟污水	ZVI	48 h	芦苇	−	88.60	92.20	−	[44]
5	潜流人工湿地	模拟污水	Fe^{3+}	96 h	鸢尾花	86.33	86.68	−	63.36	[25]
6	垂直流人工湿地	模拟污水	Fe^{2+}	16 h	水芹	86.82	78.36	−	−	[45]

注：−表示相应参考文献中未检测该指标；ZVI为零价铁。

1.3 人工湿地−微生物燃料电池

人工湿地−微生物燃料电池 (CW-MFC) 是使用生物阴极和生物阳极构建，利用微生物将化学能转化为电能的电化学系统[46]。生物电化学强化脱氮是 MFC 强化 CW 的主要方法。在 CW-MFC 中，产电微生物等功能菌在阳极处富集，发生电解反应与氧化反应，阴极位于湿地系统上层部分，便于电路性能转换从而形成稳定电流 (图 3)。产电细菌产生稳定电流，电流促进作用又可以反过来促进微生物对污染物降解[47-48]。电极材料的多孔结构有利于微生物的生长和繁殖，包括 Anammox 细菌。但是在有氧环境中，氧气可能会与氮氧化物竞争电子，因为异养需氧细菌通常会优先利用氧[49]。CW-MFC 主要通过强化系统微生物发生硝化、反硝化和 Anammox 作用，实现污水氮元素的深度去除，硝化通常发生在 CW-MFC 的阴极好氧区域，反硝化和 Anammox 位于 CW-MFC 阳极厌氧区域[50-51]。而阴极处植物根系的固氮作用对氮去除发挥一定作用，但是与生物炭和铁元素相比，CW-MFC 系统在氮素去除方面与植物联系并不十

分紧密。

CW-MFC 系统可以将废水处理效率提高 27%~49%，通过提高阴极和阳极去除污染物的效率可以进一步提高该效率，并获得更高的生物产能[52]。Wang 等[53] 研究发现，相对于对照人工湿地，CW-MFC 系统的氮去除率更高 (82.32%)(表 3)。阳极微生物群落多样性丰富，亚硝酸盐氧化菌 (微生物群落读数 144±8)，氨氧化菌 (149±7)，厌氧氨氧化菌 (281±8) 获得显著富集；将 MFC 嵌入 Anammox 复合人工湿地中，形成 Anammox-CW- MFC，可以实现生物发电、脱氮和降解有机污染物同步进行，实现清洁环境和经济效益双重目标[7,54]。然而，单纯依靠厌氧氨氧化菌去除氮素，脱氮效果并不理想，其出水含大量的硝酸盐，理论上的 TN 去除率达不到 80%[55]。因此，需要研究厌氧氨氧化细菌细胞外电子转移活性和这种偶联反应分子机制[49]。Irdis 等[56] 研究了通过控制阴极液体中溶解氧的浓度，硝化作用可以在微生物燃料电池的阴极处与反硝化作用有效地耦合，实现同步硝化与反硝化 (SND)[57-58]。但是，以上研究都未对系统产电性能进行进一步研究。另外，CW-MFC 对植物生长也有一定促进作用。Hu 等[44] 对湿地植物进行了氮同化测量，结果表明在 CW-MFC 中的湿地植物的氮同化量 (每 m^2 植物同化氮质量为 0.95 g) 大于在普通湿地中的植物氮同化量 (每 m^2 植物同化氮质量为 0.74 g)。这只是从植物生长状况显示出促进效果，是否因为电子转移尚未可知。CW-MFC 作为高效低耗处理污水新兴系统，是人工湿地中

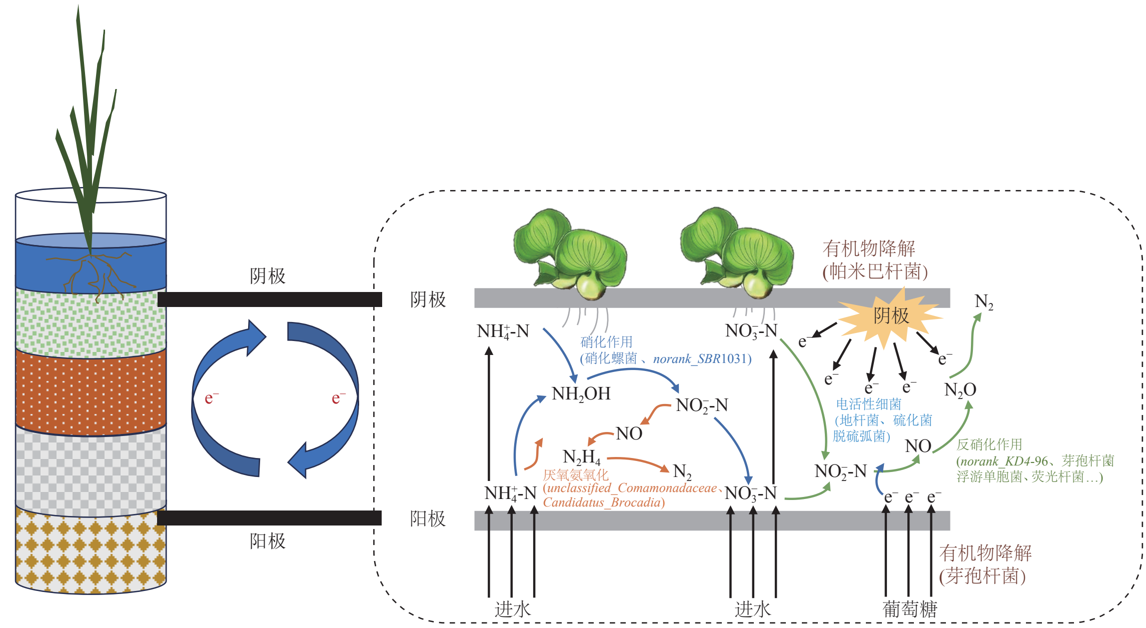

图 3 人工湿地–微生物燃料电池氮去除机制[46]

Fig.3 Mechanism of nitrogen removal in constructed wetlands-microbial fuel cells[46]

表 3 人工湿地–微生物燃料电池的污染物去除效能

Table 3 Constructed wetland-microbial fuel cell pollutant removal efficiency

序号	湿地类型	废水类型	植物	水力停留时间/d	阳极	阴极	去除效率/%				参考文献
							氨氮	总氮	总磷	化学需氧量	
1	潜流人工湿地	模拟污水	芦苇	2.6	石墨棒	石墨棒	58.00	–	–	61.00	[52]
2	垂直流人工湿地	模拟污水	香蒲	2	碳纤维毡	碳纤维毡	82.32	71.00	–	80.00	[53]
3	垂直流人工湿地	模拟污水	美人蕉	9	碳纤维	石墨板	–	76.00	–	85.00	[59]
4	潜流人工湿地	模拟污水	黄菖蒲	7	石墨毡	石墨毡	98.00	97.00	–	91.00	[60]
5	垂直流人工湿地	养猪废水	芦苇	1	石墨砾石	石墨砾石	75.00	58.00	86.00	81.00	[61]
6	垂直流人工湿地	养猪废水	美人蕉	2	不锈钢网	碳毡	75.00	–	–	88.00	[58]

注：–表示相应参考文献中未检测该指标。

有效脱氮及生物能源生产的可行解决方案，然而，由于系统受电极材料的影响显著，如何选择更廉价优质电极和基质等材料减少投入费用，是该技术应用于实际需要解决的问题。同时，CW-MFC 系统主要通过外源电能加速反应过程实现氮污染深度去除，但电能产生对植物和微生物深层影响机制，以及电子详细转移途径，由于系统复杂程度尚未得到阐明。

2 总结与展望

功能基质强化型人工湿地的污染物去除机制较为复杂，生物炭和铁元素通过优化湿地基质、强化微生物之间以及微生物与植物之间的联系，提升了系统效能；微生物燃料电池对微生物之间协作的强化作用更为显著，尽管其对植物与微生物之间的互作调控作用不明显，但可以产生电能。然而，关于这 3 种手段在人工湿地中的应用研究仍处于起步阶段，若要实现对农业氮素面源污染的有效控制，还需在以下关键领域展开深入探索：

(1) 通过利用当下人工智能技术构建生产–特性–应用关系，不仅可以通过预测和有针对性设计促进生物炭发展，而且可以提前降低生物炭在应用过程中潜在风险。

(2) 铁强化人工湿地研究主要集中在生物和非生物过程，铁元素调节系统中微生物之间、植物和功能微生物之间协作机制尚未得到有效研究。其次，铁及其载体材料生态友好性值得进一步观察。

(3)CW-MFC 对电极表面微生物作用机理的研究主要集中于微生物群落组成、丰富度和多样性的变化，但对于微生物之间，特别是产电菌与污染物降解菌之间的复杂合作与拮抗作用，以及植物与微生物之间协作机制的研究仍需进一步深入。

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Research progress on enhancing agricultural nitrogen non-point source pollution control in constructed wetlands with functional substrates

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Abstract: Agricultural nitrogen pollution has become a leading cause of non-point source pollution, resulting in widespread water quality deterioration including eutrophication, nitrate contamination of groundwater, and the formation of black and odorous water bodies. These adverse environmental impacts threaten aquatic ecosystems, public health, and hinder the advancement of rural ecological civilization. As agricultural production intensifies and expands, especially in rural areas lacking centralized treatment infrastructure, developing effective and sustainable nitrogen removal technologies is imperative for environmental protection.

Constructed wetlands (CW) have been widely adopted as cost-effective, eco-friendly wastewater treatment systems that integrate physical, chemical, and biological processes for pollutant removal. Their low operational cost, landscape compatibility, and ability to support plant-microbe interactions make them particularly suitable for decentralized rural wastewater treatment. However, traditional CW often exhibit limited nitrogen removal efficiency and instability due to variable hydraulic loading, seasonal temperature fluctuations, and low carbon-to-nitrogen (C/N) ratios, which impede microbial nitrogen transformation processes. This review focuses on overcoming the challenges of nitrogen removal in CW by integrating functional substrates, including biochar, iron-based materials, and microbial fuel cells (MFC). These substrates enhance CW performance by improving substrate physicochemical properties, stimulating microbial communities, and facilitating nitrogen transformation under diverse environmental conditions. Biochar, produced by pyrolysis of biomass, is a porous carbonaceous material with high surface area and abundant functional groups, promoting pollutant adsorption and providing habitats for microorganisms. It improves redox potential and simultaneously supports aerobic nitrification and anaerobic denitrification. Iron-based materials, such as zero-valent iron and ferrous oxides, play crucial roles in redox reactions, facilitating denitrification and anaerobic ammonium oxidation (anammox). They also aid phosphorus adsorption and immobilize heavy metals, improving overall water quality. Combined application of biochar and iron synergistically enhances substrate stability and microbial diversity, promoting efficient nitrogen cycling and stronger plant-microbe interactions. MFC introduce bioelectrochemical functions by enabling extracellular electron transfer and electricity generation through microbial metabolism. They increase nitrogen removal efficiency, especially under low carbon availability, by stimulating microbial cooperation and redox activity. Though MFC effects on plant growth are limited, they provide added value by generating renewable energy to support system operation in off-grid rural settings. Furthermore, the use of these functional substrates helps to mitigate greenhouse gas emissions commonly associated with nitrogen cycling in wetlands, thereby contributing to climate change mitigation efforts. Their incorporation into CW design not only improves nitrogen removal but also enhances the overall ecological sustainability of treatment systems. Recent advances have demonstrated that combining these substrates with optimized operational parameters can significantly extend the lifespan and effectiveness of constructed wetlands, reduce maintenance costs and improve resilience to environmental fluctuations. his review synthesizes recent advances in the use of functional substrates for CW enhancement, elucidating their mechanisms, advantages under stress conditions, and practical implementation strategies. The integration of biochar, iron, and MFC offers a comprehensive and innovative approach to mitigate agricultural non-point source nitrogen pollution. Ultimately, this strategy improves nitrogen removal efficiency, system resilience, and sustainability, contributing significantly to water resource protection and rural ecological civilization development across diverse geographical regions worldwide with sustainable way.

Keywords: non-point source nitrogen pollution; novel constructed wetlands; functional substrate; black and odorous water