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水产养殖尾水除磷技术研究进展*

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摘要 磷是构成生命体的必需元素, 在生物生长过程中发挥着重要作用, 然而水体中磷元素浓度超标易导致水体富营养化和蓝藻水华等问题, 影响生态环境和经济发展。在“大食物观”背景下, 水产养殖的绿色发展尤为重要, 如何妥善处理养殖尾水中高浓度氮、磷营养盐, 实现尾水磷达标排放或资源化利用已引起广泛重视。本文首先通过 VOSviewer 文献可视化软件的关键词贡献分析了近 10 年水产养殖尾水除磷技术的研究现状, 详细介绍了水体中磷的赋存形态分类, 阐述了国内外水产养殖尾水除磷技术分类、工作原理和研究进展, 分析各个除磷技术的优缺点; 同时针对现行水产养殖工艺的尾水磷处理方法进行了总结与展望。本文统领分析了水产养殖尾水除磷原理及工艺技术, 可为尾水磷处理和资源回收提供新思路, 推动水产养殖可持续发展。

关键词 磷; 赋存形态; 水产养殖尾水; 循环水养殖; 除磷方法

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磷是构成动植物等生命体的必需元素, 水体缺磷会限制初级生产力, 磷作为不可再生资源, 其短缺问题引起国际重视(Huang *et al*, 2021); 然而磷浓度过高可导致水体富营养化, 影响人类生产生活、危害水体健康, 造成严重经济损失(Masatoshi *et al*, 2023; Zhang A *et al*, 2024), 因此, 水体中多余磷去除和磷资源回收的研究具有重要意义。

在“大食物观”背景下, 水产养殖业高质量发展已成为我国基本战略。根据《2024 中国渔业统计年鉴》报告显示, 我国 2023 年水产品总产量为 7 116.17 万 t, 同比增长 4.39%, 其中养殖产量占比 81.6%。根据 2024 年中国生态环境状况公报, 2023 年我国内陆江河重要渔业水域总磷超标面积占 27.3%; 海洋天然重要渔业水域中活性磷酸盐超标面积为 27.0%, 海水重

点养殖区中活性磷酸盐超标面积为 28.2%。我国海水养殖每年向近海排入的总氮、总磷分别为 4.77×10^4 t 和 3.75×10^3 t (2022 年中国生态环境状况公报; 2023 年中国生态环境状况公报)。2022—2024 年我国各省市养殖尾水排放标准陆续出台, 以山东省为例, DB374676-2023《海水养殖尾水排放标准》提出总磷一级排放限值 0.7 mg/L, 二级排放限值 1.0 mg/L, 如何实现养殖尾水中磷高效去除、甚至回收已成为支持我国水产养殖业绿色发展的重点之一。

研究论文的发表情况可以在一定程度上反映该领域的关注热点和发展方向, 如图 1 所示, 近 10 年 CNKI 关于“水产养殖”“磷去除”关键词发文量约 78 篇, 图 2 为近 10 年 Web of Science 关于“水产养殖”“除磷”关键词发文量约 214 篇, 通过运用 VOSviewer 可视

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溶解态磷(dissolved phosphorus, DP)和颗粒态磷(particulate phosphorus, PP)(Hernández-Ramírez *et al.*, 2023)。按照化学态可将水体中磷分成无机磷(inorganic phosphorus, IP)和有机磷(organic phosphorus, OP)。DP分为可溶态有机磷(dissolved organic phosphorus, DOP)和可溶态无机磷(dissolved inorganic phosphorus, DIP)(吴怡等, 2010), 也分为可溶活性态磷(soluble reactive phosphorus, SRP)和可溶非活性态磷(soluble unreactive phosphorus, SUP)(Cooper *et al.*, 2002)。DIP包括缩合磷酸盐(condensed phosphorus 或 dissolved acid hydrolysable phosphorus, DAHP)和 SRP(Tue-Ngeun *et al.*, 2005)。SUP包含 DOP和 DAHP(Evans *et al.*, 2004)。DAHP分为焦磷酸盐、多聚磷酸盐和偏磷酸盐等(Tue-Ngeun *et al.*, 2005)。颗粒态磷(PP)指存在于水体悬浮颗粒物表面或内部的磷(Loh *et al.*, 2024)。PP可分为颗粒态有机磷(particulate organic phosphorus, POP)和颗粒态无机磷(particulate inorganic phosphorus, PIP)(Feng *et al.*, 2024)。值得关注的是 SRP是植物直接吸收利用的磷酸盐形式, 当前我国绝大部分水产养殖尾水将水体 TP及 SRP规定为限制排放标准(何敏等, 2023)。

2 水产养殖尾水除磷技术

国内外专家持续推动水产养殖尾水除磷技术研究, 基于水体除磷技术的原理, 集成物理、化学、生物等技术, 更好地将磷去除方法应用于不同水产养殖模式尾水治理中; 同时, 磷作为不可再生资源, 养殖尾水中的磷回收逐渐引起重视。

2.1 水产养殖尾水除磷技术原理

2.1.1 物理法除磷技术

(1) 吸附法

吸附法除磷是指吸附剂通过离子交换、配位络合、静电吸附和表面吸附等机制, 去除污水中磷的方法(仇付国等, 2023)。吸附材料具有适应性广、吸附速度快、可重复利用的优点, 但此法吸附饱和量低, 受 pH影响较大, 在中性时很难去除磷。由于吸附剂吸附能力与其比表面积、孔隙率和表面官能团密切相关, 因此吸附剂的种类与性质是影响磷净化效果的主要因素。Tan 等(2022)学者在实际水产养殖尾水中加入煅烧蛋壳吸附硝态氮和磷酸盐($\text{PO}_4^{3-}\text{-P}$), 发现 $\text{PO}_4^{3-}\text{-P}$ 去除率为 97.3%。另外, 吸附过程中, 随着时间的延长, 吸附剂会趋于饱和, 必须不断补充新吸附剂以确保使用效果。研制吸附能力强、吸附容量大且可再生的吸附剂是关键。

(2) 膜分离磷技术

膜分离技术是通过膜的选择性, 用膜两侧的浓度差、液压差和电位差等差值作为动力, 将养殖尾水污染物截留完成污染物分离, 属物理分离, 常见的膜分离技术有微滤、纳滤、超滤、反渗透以及液膜等手段(陈铭真, 2022)。由于膜的分离效果与膜材料、孔径及表面特性等密切相关, 因此选择适宜的膜对保障分离效果至关重要。Teoh 等(2022)分别使用聚砜膜和聚偏氟乙烯膜净化养殖鲶鱼(*Silurus*)的尾水, 出水 TP 低于 0.02 mg/L。随着污染物在膜表面的积累, 易造成膜污染而影响分离效果, 也导致膜寿命缩短, 因此, 膜抗污染和再生等性能是该技术的关键(Zhou *et al.*, 2021; Law *et al.*, 2018)。膜分离技术也可与化学沉淀法、生物反应器结合形成膜组合工艺, 提高尾水中磷的去除效率(李媛等, 2024)。

2.1.2 化学法除磷技术

(1) 沉淀法

沉淀法除磷是向水体中投加化学药剂, Fe^{2+} 或 Fe^{3+} 与磷酸盐生成不规则的磷酸盐固相沉淀, 再通过絮凝、固液分离技术去除磷的一种方法, 主要受 2 个因素影响: 体系 pH 和磷赋存形态(Xu *et al.*, 2024; 孙明玮, 2018)。该方法使用的除磷剂一般有: 镁盐、碳酸盐、钙盐、铝盐和铁盐等金属类物质(Lei *et al.*, 2021; 倪琳琳等, 2024)。在使用铝盐除磷的过程中要严加控制 pH, 因为 Al^{3+} 具有两性特性, 体系的 pH 不宜过高, 否则会降低磷的去除效果(王广伟等, 2010)。张旺等(2023)以淡水池塘养殖水体为研究对象, 比较了明矾和聚硫酸铁对养殖水体总磷的去除效果, 其中明矾对总磷的去除率(90.10%)高于聚硫酸铁的去除率(88.74%)。化学沉淀除磷技术应用虽广泛, 但也有缺点。金虎等(2019)使用聚合氯化铝强化 A^2/O 工艺生物除磷时, 发现副产物会导致生物除磷能力下降。此外, 该方法需要大量药剂, 一是成本高; 二是产生大量化学污泥, 且强化生物除磷时, 化学沉淀法较难去除低浓度磷(如低于 0.1 mg P/L)(Lei *et al.*, 2021)。

(2) 结晶法

结晶法除磷是在过饱和状态下, Mg^{2+} 、 Ca^{2+} 、 Fe^{2+} 或 Fe^{3+} 等与磷酸盐发生反应, 自发形成晶核或以固相微粒充当晶核, 持续长成规则的磷晶体, 最后从溶液中分离出来, 实现磷的去除与回收(Guo *et al.*, 2020; Agrawal *et al.*, 2021), 与沉淀法的区别在于晶体有固定的熔点和沸点。结晶法可分为 3 种: 磷酸铵镁法(Zhang *et al.*, 2016)、羟基磷酸钙法(Azis *et al.*, 2018)和蓝铁矿 [$\text{Fe}_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$] 法(Bonisławska *et al.*,

2019)。离子摩尔比、共存离子、晶种类型等是影响结晶法的主要因素(Amin *et al*, 2024)。Zhang 等(2016)利用磷酸铵镁结晶法除磷,模拟海水循环水养殖尾水处理,明显降低了尾水磷浓度,且鸟粪石结晶的平均尺寸随着 pH 的增大而增大。

(3) 电化学絮凝法

电化学絮凝是在外电场作用下,金属阳极氧化生成金属阳离子,经过水解和聚合生成具有凝聚和吸附

作用的氢氧化物,以此去除水中污染物,因为不引入其他化学物质,电化学絮凝是一种环境友好型的处理方法(张旺等, 2023)。徐进等(2014)研究表明,电絮凝产生的多核羟基化合物对水中磷物质有较好的吸附效果,具有高效安全、反应速度快、易操作等特点。当利用铁、铝等阳极材料时,阳极电解生成金属离子及其化合物,与水中的无机磷生成磷酸盐沉淀,其原理如表 1 所示。

表 1 电絮凝铝、铁阳极去除磷机理

Tab.1 Mechanism of phosphorus removal by electro flocculating aluminums and iron anodes

电极 Electrodes	阳极反应 Anodic reaction	阴极反应 Cathodic reaction	溶液反应 Solution reaction
铝电极 Aluminum electrode	$2Al \rightarrow 2Al^{3+} + 6e$	$3H_2O + 3e \rightarrow 3/2H_2\uparrow + 3OH^-$	$Al^{3+} + 3H_2O \rightarrow Al(OH)_3\downarrow + 3H^+$ $Al^{3+} + PO_4^{3-} \rightarrow AlPO_4\downarrow$
铁电极 Iron electrode	$4Fe \rightarrow 4Fe^{2+} + 8e$	$2H^+ + 2e \rightarrow H_2\uparrow$	$Fe^{2+} + 4H^+ + O_2 \rightarrow 2Fe^{3+} + 2H_2O$ $Fe^{3+} + PO_4^{3-} \rightarrow FePO_4\downarrow$ $3Fe^{2+} + 2PO_4^{3-} \rightarrow Fe_3(PO_4)_2\downarrow$

(4) 深度氧化法(oxidation processes, AOPs)

深度氧化法除磷主要是借助光、氧化剂及催化剂等产生羟基自由基,使尾水氮、磷及生物分子发生氧化降解或衰亡,达到净化作用,根据氧化剂类型可分为光(紫外)、臭氧和芬顿等氧化法(孟锋等, 2020)。Gomes 等(2020)使用芬顿氧化法处理尼罗河鲮鱼(*Oreochromis niloticus*)养殖尾水,发现出水 PO_4^{3-} -P 浓度降低至 0.14 mg/L 以下。目前, AOPs 技术仍存在一定的局限性,因为氧化剂如 O_3 、 H_2O_2 等可能会影响水产动物生长(Gomes *et al*, 2020),同时,自由基非选择性氧化作用可能会产生有毒有害物质造成二次污染(Gorito *et al*, 2022),故需进一步评估 AOPs 的安全性。

2.1.3 生物法除磷技术

(1) 生物反应器除磷技术

生物反应器是目前养殖尾水处理的常用方法,是一种利用附着在反应器填料表面的活性生物(如藻类、微生物)进行生物反应的处理方法,分离净化养殖尾水中氮磷等污染物。传统生物除磷是聚磷菌在好氧时,不断氧化体内的 β -羟基丁酸(PHB),从外界摄取水体中的磷,将磷以聚磷酸盐的形式储存在细胞内;在厌氧时,聚磷菌释放体内聚磷酸盐,吸收废水中的有机物,从而合成 PHB(邢超等, 2022)。反硝化除磷是反硝化聚磷菌(denitrifying phosphate accumulating organism, DPAO)在厌氧时释磷;缺氧时,反硝化聚磷菌采用电子受体(NO_2 和 NO_3)氧化 PHB 以产生能量,从环境中吸收磷(Marques *et al*, 2017; 易鑫等, 2024)。这二者的区别在于聚磷菌的电子受体只能

为氧,反硝化聚磷菌的电子受体可以是亚硝酸盐和硝酸盐。目前的生物反应器类型主要有移动床生物反应器(MBBR)和固定床生物反应器(FBBR)。MBBR 既具有活性污泥法的高效性和运转灵活性,又具有传统生物膜法耐冲击负荷、污泥龄长、剩余污泥少的特点,它结合了传统流化床和生物接触氧化的优点,解决了 FBBR 需要定期反冲洗,流化床需要将载体流化,淹没式生物滤池易堵塞,需要清洗填料更换曝气器等问题。目前,膜式光生物反应器即生物膜与微藻耦合成藻类生物膜进行水产养殖尾水处理,逐渐引起学者重视, Zhang H C 等(2024)采用旋转藻类生物膜对海水养殖尾水净化处理,结果表明,该系统是处理海水养殖尾水的有效设备,其中磷酸盐去除率高达 99%。目前使用较广泛的生物反应器类型及应用情况见表 2。

(2) 生物絮团(biofloc technology, BFT)除磷技术

BFT 除磷主要是利用异养细菌,如反硝化聚磷菌等细菌将尾水中的磷物质消耗,并通过自身细胞或代谢物的絮凝作用将固体微粒絮凝成大颗粒,再借助固液分离技术达到净化目的。C/N 比是影响生物絮团技术的关键因素。陈伟等(2018)探究不同 C/N 比对花鳃鲰(*Anguilla marmorata*)循环水养殖系统生物絮凝反应器脱氮除磷的影响,得出 C/N 升高,反应器脱氮除磷效果也逐渐增加, C/N=15 时去除效果最佳, TN、 NO_3 和 PO_4^{3-} 的去除率分别为 46.60%、43.49%和 24.40%。研究表明,生物絮团浓度控制要适宜,浓度过低预期效果差,过高可能引起鱼虾的鳃堵塞导致呼吸困难(Ogello *et al*, 2021; Robles *et al*, 2020)。当前,生物絮

团除磷效果并不理想,因此,研究同步高效脱氮除磷的菌株具有重要意义(Figueroa-Espinoza *et al*, 2022)。

(3)微藻生物净化除磷技术

磷是微藻生长的基本元素,是合成核酸、磷脂和叶绿素所必需的。微藻是一种光合自养型微生物,具有繁殖速度快、环境适应能力强和生长周期短等特性,可以去除尾水中氮、磷等污染物(Nogueira *et al*, 2018; Zhang *et al*, 2023)。宋楚儿(2023)将异养蛋白核小球藻(*Chlorella vulgaris*)应用于对虾海水养殖尾水

处理技术中,研究得出,蛋白核小球藻能明显降低养殖尾水中的磷,且同步脱氮,磷酸盐去除率随着小球藻细胞生长持续上升,去除率达(81.40±9.86)%。王美琦等(2022)研究表明,蛋白核小球藻对 DOP 的去除机制包括 2 个方面:一是直接吸收的小分子 DOP,可通过微藻细胞膜上的相关转运蛋白主动转运到细胞质中。二是水解后吸收的 DOP 被水解酶水解成无机磷,以磷酸盐的形式被吸收。如图 4 所示,微藻可通过自身生长、化学沉淀和氧气悬浮、表面吸附的作

表 2 生物反应器除磷技术的应用情况及优缺点

Tab.2 Application of bioreactor phosphorus removal technology and its advantages and disadvantages

生物反应器 Bioreactor	技术原理及应用情况 Principles and current situation	优点 Advantage	缺点 Disadvantage	参考文献 Reference
动态膜生物反应器 Dynamic membrane bioreactor	利用尾水在通过基材时所截留下来的污泥颗粒形成生物膜,目前在海水养殖尾水脱氮除磷上多处于试验阶段。	节能减排,降低尾水处理成本。	基材需要长期维护,尾水盐度较高时处理效果较差。	高教成, 2016; 吴梦等, 2019
曝气膜生物反应器 Aeration membrane bioreactor	通过中空纤维膜进行无泡曝气,形成生物膜分层,实现硝化反硝化脱氮除磷,多用于去除尾水中的难降解有机物和挥发性有机污染物。	可同时实现硝化反硝化,高效,占地面积小。	中空纤维膜的维护和密闭稳定性差,供氧效率低。	Hong <i>et al</i> , 2012; Chen <i>et al</i> , 2023
序批式膜生物反应器 Sequencing batch membrane bioreactor	厌氧/缺氧/好氧(A ² /O)的工艺结合 MBR,多用于海水养殖尾水脱氮除磷。	脱氮除磷效率高,膜污染较轻。	填料选择困难,需额外添加碳源。	Chen <i>et al</i> , 2021; Lu <i>et al</i> , 2021
膜式光生物反应器 Membrane photobioreactor	将膜与微藻结合,结合微藻的光合作用,可用于海水养殖尾水脱氮除磷。	膜污染较轻,脱氮除磷效率高	膜技术和运行维护复杂。	Tao <i>et al</i> , 2021; Tian <i>et al</i> , 2020

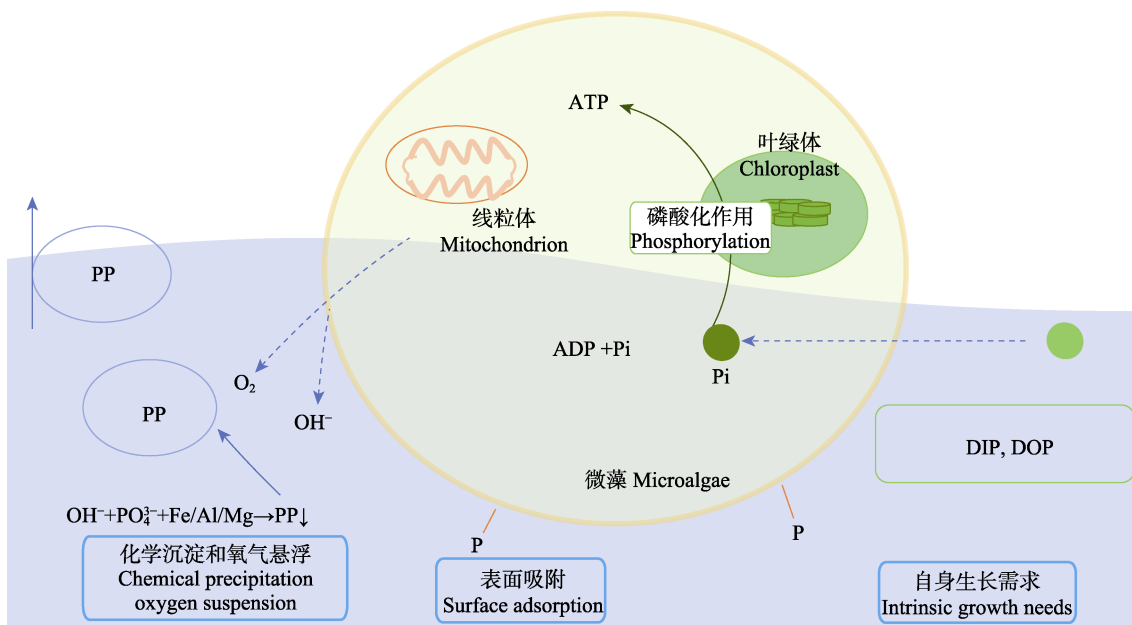


图 4 微藻对水体磷的去除机制: 磷酸化作用、化学沉淀和氧气悬浮、表面吸附、自身生长需求

Fig.4 Mechanisms of phosphorus removal from water bodies by microalgae including phosphorylation, chemical precipitation, oxygen suspension, surface adsorption and intrinsic growth needs

用去除水体中的磷(安余梁等, 2024)。因此, 光合作用对于微藻去除磷至关重要, 光照、微藻种类与数量、氮磷比和环境温度对微藻生物净化除磷均有影响。目前, 菱形藻(*Nitzschia* sp.)、小球藻和栅藻(*Scenedesmus*)用于养殖尾水除磷, 张秀霞等(2024)研究 5 种微藻处理对虾池塘养殖尾水, 结果表明, 菱形藻对 TP 的去除效率最高(71.43%), 其他微藻的 TP 去除率均为 40% 以上。Liu 等(2019)使用四尾栅藻(*Scenedesmus* sp.)处理水产养殖尾水, 出水 TP 为 0.18 mg/L, 符合养殖尾水排放要求。因此, 针对不同的养殖情况和水体盐度, 应选择合适的微藻进行养殖尾水除磷, 但如何高效收集微藻仍在制约该技术在尾水处理中的应用。

(4) 菌藻协同法除磷技术

菌藻共生系统的提出是为了取代活性污泥改进传统藻类塘, 提高脱氮除磷效率。菌藻共生是将微藻与微生物制剂有机结合在一起净化污水, 作用机理是利用微藻的光合作用向水体输送 O₂, 供给好氧异养型微生物进行代谢活动, 增强好氧异养微生物对有机污染物氧化分解的能力; 同时, 好氧异养微生物的代

谢产物(如无机氮和磷化合物)或呼吸产物(如 CO₂)可为藻类进行光合作用提供碳源(金忠友等, 2023), 其中, 微生物制剂除磷机理为: 聚磷菌厌氧释磷、好氧吸磷, 除磷效果取决于聚磷菌在厌氧阶段的释磷量(白瑞等, 2017), 而现阶段多采用反硝化聚磷菌除磷。Gao 等(2022)构建了一种集产酸发酵与微藻培养于一体的菌藻协同反应器(BACR), 并探究了该反应器在处理海水养殖尾水方面的可行性。结果表明, BACR 防止了微藻被发酵细菌污染, 促进了微藻生长, 增强了 BACR 对海水养殖尾水中 TP 的有效去除。Ding 等(2020)在室温下构建具有内部循环流化床(ICFB)的微藻-膜生物反应器(MMBR), 以研究海水养殖尾水污染物的去除效率, 运行 40 d 内, 磷酸盐去除率达到 80%。Babatsouli 等(2015)研究了固定床生物反应器-微藻系统处理海水养殖尾水, 结果表明, 磷酸盐的去除与有机物浓度具有相关性, 当有机物浓度较高时, 磷酸盐去除率较高。当前菌藻共生系统可分为悬浮型菌藻共生系统和固定型菌藻共生系统, 见表 3。

表 3 菌藻共生系统的应用情况及优缺点

Tab.3 Application of bacterial-algae symbiosis system and its advantages and disadvantages

菌藻共生系统 Symbiotic system of bacteria and algae	应用形式 Application form	应用情况 Application situation	优点 Advantage	缺点 Disadvantage	参考文献 Reference
悬浮型菌藻共生系统 Suspended bacterial-algae symbiotic system	利用细菌和微藻在废水中的分散状态, 相互作用实现污染物的去除。	一般在传统稳定塘的基础上添加菌藻, 形成高效藻类塘。	污染物处理效率高, 藻类沉降性能优异。	处理效果不稳定, 系统产泥量大, 需定期排泥。	王荣昌等, 2018
固定型菌藻共生系统 Immobilized bacterial-algae symbiotic system	采用高分子材料(如海藻酸盐、聚乙烯醇等)或真菌载体对菌藻进行吸附、包埋被动固定于特定空间。	应用范围较广, 一般应用于水产养殖尾水二级出水脱氮除磷。	系统产泥量少, 可重复利用, 系统稳定性强。	易出现脱落现象, 成本偏高, 制备程序较为复杂。	马洪婧等, 2022; 王荣昌等, 2018
菌藻共生生物膜系统 Bacterial-algae symbiotic system	属于固定型菌藻共生系统, 是利用细菌和微藻附着在载体表面, 通过自身胞外聚合物主动胶连在一起。	一般应用于生物膜反应器中除磷, 如间歇式生物膜反应器、序批式生物膜反应器等。	系统产泥量少, 价格低廉, 处理效率高。	生物膜易脱落、膜污染, 系统稳定性较低。	Zheng <i>et al.</i> , 2021

(5) 人工湿地法除磷技术

人工湿地是人为将基质、微生物和植物按一定比例组成的人工生态系统, 结合自然界中物理、化学和生物的共同作用实现污水处理和净化(张冉等, 2016)。人工湿地主要包括 3 种: 表面流人工湿地、潜流人工湿地、垂直潜流人工湿地(Ruppelt *et al.*, 2018)。

人工湿地的除磷机制分为吸附、吸收和沉淀 3 种

(Verma *et al.*, 2022)。人工湿地吸附除磷是通过比表面积较大、多孔吸附剂作为填料实现的(谭心等, 2021)。因此, 填料的选择尤为重要, 需考虑填料比表面积、多孔性、可变电荷位点、表面官能团和改性方法等。Shang 等(2022)将新型的载钼氨化水热生物炭作为人工湿地填料基质, 其磷吸附容量较高(43.1mg/g)。同时, 在人工湿地基础上可结合化学沉淀技术将水中磷

离子转为不溶性盐沉淀物(Kasak *et al.*, 2020)。Zeng 等(2020)研究镁催化剂对湿地磷回收效果的影响, 发现加入 MgO 后, OP 回收率从 5.25%升到 93.42%, 当 pH=10 时, 磷回收率达 98.47%。Leng 等(2023)研究以石榴石为基质的人工湿地耦合微生物燃料电池系统(CW-MFC), 该系统对磷的去除机制主要是石榴石组分扩散到污水中, 与游离 $\text{PO}_4^{3-}\text{-P}$ 和 HPO_4^{2-} 发生反应, 形成 $\text{Mg}_4(\text{PO}_4)_2\text{OH}$ 、 $\text{AlPO}_4(\text{H}_2\text{O})_{1.5}$ 和 CaPO_4 沉淀物, TP 去除率达 92.4%。同时, CW-MFC 和曝气技术联合用于处理低碳氮比和高盐度的海水养殖尾水, Wang 等(2023)采用 CW-MFC 和虹吸曝气技术, 微生物染料电池在电活性细菌基础上, 耦合了藻类生物膜处理水质, 结果表明, 该系统对各营养盐均有明显去除作用, 总磷去除率高达(92.59±3.13)%。

2.2 现行水产养殖工艺尾水除磷技术

为追求更好的除磷效率和提高水产养殖可持续发展水平, 根据不同水产养殖模式特点, 采取不同的水产养殖尾水除磷方法, 通常结合物理、化学和生物等方法进行尾水除磷和磷资源回收。

2.2.1 循环水养殖尾水除磷技术 工厂化循环水养殖(RAS)养殖密度高、饲料投喂量大, 导致营养物质积累, 水体中氮磷浓度较高。Yogev 等(2020)建立了一种近乎零排放的 RAS, 由养鱼池、好氧(硝化生物反应器)、缺氧(反硝化生物反应器)和厌氧反应器 4 个部分组成, 厌氧处理回路为上流式厌氧污泥层反应器(UASB), 用于处理硝化反硝化产生的富磷污泥, 经 UASB 处理后, 污泥被矿化为磷灰石和鸟粪石。伍建业等(2023)以“高效生态浮床-垂直流人工湿地-沉水植物池单元”的三级单元复合人工湿地为研究对象, 对复合人工湿地系统中设置挂膜, 研究不同干湿交替时间下该系统对高密度陆基水产养殖尾水氮、磷的净化效果。结果表明, 干湿比为 16 h : 8 h 时, 复合人工湿地系统对 TP 的平均去除率为 56.85%~77.50%, 对磷的去除主要发生在二级单元即垂直流人工湿地, 这可能与二级单元的填料(气泡砖和赤泥)中含有铁、铝、钙等离子有关, PO_4^{3-} 可与这些离子结合形成溶解度较低的 FePO_4 、 AlPO_4 及不溶性的 $\text{Ca}_3(\text{PO}_4)_2$ 。因此, 提高过滤介质的磷结合能力是增强磷去除效果的最佳策略(李丹等, 2020)。近年来, RAS 系统与多营养层次综合养殖系统(integrated multi-trophic aquaculture, IMTA)的结合, 亦可实现高效的尾水除磷效果。IMTA 是指利用各营养级水生动植物在食物链、生态位等方面互惠互利的原理, 合理调配各营养层级的空间分布, 实现饵料优化配置和代谢物综合利用, 从源头减

少氮磷等污染物的产生。Li 等(2019)将欧洲舌齿鲈(*Dicentrarchus labrax*)RAS、高速率微藻池(high rate algal ponds, HRAP)和牡蛎池结合, 组成 IMTA 系统, HRAP 对鱼池出水进行生物修复, 牡蛎摄入微藻, 结果表明, IMTA 系统对氨氮、硝酸盐、亚硝酸盐和磷酸盐的去除率都超过了 96%。

循环水养殖高效一体化尾水处理系统的研究逐渐引起学者重视, 我国早在 2006 年提出水产养殖有机污水一体化净化设备的技术理念, 杨菁等(2006)将超声波及纳米电气石功能材料有机结合, 建立新颖滤池型式, 并且整机采用可移式一体化设计, 包括气提组件、物理过滤组件、纳米滤料-超声波生物滤池、发生器、可移式机架等组件。形成了工艺简化、结构紧凑、处理水质稳定、使用方便的一体化纳米过滤净水设备, 对氮磷均有一定的去除效果。商洪国等(2021)采用混合营养型, 即钝顶螺旋藻(*Spirulina platensis*)结合膜光合反应器考察碳氮比对海水鲑鱼循环水养殖中碳、氮、磷一体化去除效率和藻生物产量的影响, 结果表明, 钝顶螺旋藻可实现碳、氮、磷高效一体化去除, 总磷的去除率高达 93%。一体化设备在当前的研究仍属于试验阶段, 在淡水养殖中应用较多, 由于海水养殖的高盐分和低碳氮比的原因, 海水养殖一体化设备的应用仍存在一定的局限性。

2.2.2 鱼菜共生养殖尾水除磷技术 鱼菜共生将养殖尾水与水培种植进行有机整合, 以高氮磷养殖尾水作为肥料供蔬菜生长, 同时利用附着微生物和蔬菜吸收氮磷完成尾水回用, 实现鱼类、植物生态共生。因此, 鱼菜种类和微生物附生载体的精配是确保净化效果和提高系统产出的根本。闫玉杰等(2022)比较了蕹菜(*Ipomoea aquatica*)、芹菜(*Apium graveolens*)、番茄(*Solanum lycopersicum*)和黄瓜(*Cucumis sativus*)对循环鱼菜共生系统中水质的净化效果, 结果表明, 蕹菜对水体中氮和总磷的去除率分别达到 72.63%和 55.62%, 去除效果最佳。鲍婷(2021)对小白菜-黄颡鱼共作系统中氮磷利用率研究的结果中显示, 种植小白菜后, 对磷利用率提高 36.06%。

鱼菜共生养殖模式多用于淡水鱼类养殖, 如尼罗罗非鱼(*Oreochromis niloticus*)、鲢鱼(*Hypophthalmichthys molitrix*)和鳙鱼等(Yep *et al.*, 2019; 朱建辉等, 2021), 因为海水盐度较高, 耐盐经济作物较少, 鱼菜共生在海水养殖方面多处于试验阶段。Alarcón-Silvas 等(2021)研究凡纳对虾(*Penaeus vannamei*)低盐度的循环水养殖, 构建了 3 个单元(水产养殖、水调节和水培), 结果表明, 该系统可减少井水中 23%的总磷和低盐度海水中 18%的总磷。

2.2.3 池塘养殖尾水除磷技术 池塘养殖是水产养殖业最基本的养殖模式。张俊等(2024)研究表明,池塘养殖投喂的饲料中有 5%~10%未被养殖对象吸收,以残饵的形式沉淀,而被养殖对象摄食的饲料中又有 25%~30%不能被吸收,而是以粪便的形式直接排出。现有的池塘养殖尾水处理多采用生物净化为主,物理化学净化为辅的方式,例如“三池两坝”、人工湿地、鱼菜共生等方法。王磊等(2023)研究“三池两坝”多级池塘处理凡纳对虾海水养殖尾水,结果表明,DIP 去除率为(85.7±9.73)%; TP 去除率为(48.3±5.75)%。阙祥尧等(2024)比较了蕹菜、凤眼莲(*Eichhornia crassipes*)、喜旱莲子草(*Alternanthera philoxeroides*) 3 种水生植物潜流人工湿地,模拟处理草鱼(*Ctenopharyngodon idellus*)池塘养殖尾水,结果表明,蕹菜对总磷的去除率最高(24.7%)。Ju 等(2015)构建了柄海鞘-微藻-刺参混养系统,结果显示,出水 TN 和 TP 浓度均降低,同时,柄海鞘(*Styela clava*)、刺参(*Apostichopus japonicas*)存活率均超过 75%。de Morais 等(2023)将凡纳对虾-罗非鱼-海藻作为 IMTA 系统,其中海藻作为生物絮凝生产系统中的无机消费者,探究不同海藻密度下,养殖尾水水质变化,其中在海藻密度为 1 g/L 时,磷回收率最高(34%)。

3 总结与展望

水产养殖尾水除磷技术的发展得益于各个领域研究技术的有机融合和不断扩展。当前,尾水除磷研究主要集中在如何实现同步高效脱氮除磷,聚焦于包括生物膜反应器、人工湿地技术、微藻生物净化和菌藻协同技术(固定化菌藻技术、菌藻生物膜技术)等生物方法。生物膜反应器除磷研究主要集中在聚磷菌和反硝化聚磷菌的培养与筛选,其中水力停留时间、盐度和溶解氧被认为是影响生物膜反应器运行效果的重要参数。人工湿地除磷技术被认为是保障水产养殖可持续发展的重要技术,除了人工湿地类型的选择、植物的选取、填料的改善、水力停留时间及水力负荷的设计及相应研究外,人工湿地技术也与电子供给新模式相结合,例如人工湿地与微生物燃料电池、沉积物燃料电池等技术的结合,从而不断提升水产养殖尾水中磷去除和回收效率。

中国作为水产养殖大国,养殖产量逐年增长,保证优质水产品供应的同时,需以“生态优先”为前提,2025 年我国也将初步实现工厂化养殖尾水的自行监测。为推动水产养殖可持续发展,高效去除养殖尾水中的磷至关重要,应“因水制宜”选择合适的水体除磷

方法,充分考虑养殖地理区域、经济发展、养殖对象排泄特征等条件及因素,融合各除磷方法的优势,取长补短,实现养殖尾水达标排放和磷回收的双重目标。为了获得更加稳定高效的除磷及回收效果,今后需基于不同养殖模式特征,开展以下深入研究:(1)水产养殖固体废弃物的高效处理及资源化利用,实现颗粒磷的回收。水产养殖过程中产生的固体废弃物如:残饵、粪便、虾蟹壳等,养殖固废是水体氮磷等污染物的源与汇,因此,针对养殖固废的分离、收集、处理和资源化利用的研究具有重要意义。在当前研究中,Wu 等(2022)利用热裂解技术将海水鱼残饵粪便制备为生物炭,再将生物炭作为污染物去除剂改良水体或土壤,进而实现养殖固体废弃物的资源化利用;(2)进一步优化生物法除磷技术,分别从效果、成本方面进一步优化技术,例如生物膜反应器、菌藻协同技术,提高其膜持久高效、易处理回收的特性。并且充分将电化学除磷技术与生物法除磷技术相结合,利用电化学技术的高效清洁特点。在外加电场的电化学除磷技术方面,可探究将清洁能源作为其驱动生产力,在自身电场的电化学除磷技术方面,优化电极材料和反应器装置,进一步探究更稳定且高效持久的阳极材料,进而提高尾水磷去除和回收效率;(3)生物工程技术改造或探究生长特性优异的耐盐碱植物,以扩大海水养殖尾水处理中植物的应用范围,深入了解海水养殖尾水处理与生长特性优异的耐盐碱植物之间的微生物互作关系,进而缓解鱼菜共生、人工湿地等生物生态法除磷技术在海水养殖尾水除磷技术方面存在的局限性。

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Research Progress on Water Body Phosphorus Removal Technology Based on Aquaculture Tail Water Treatment

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Abstract Phosphorus is an essential element for plants, animals, and other living organisms. The lack of phosphorus in aquatic environments can restrict primary productivity, a concern that has increasingly attracted global attention. However, the high phosphorus concentration leads to the eutrophication of water bodies, impacting human activities, compromising water quality, and causing notable economic losses. Therefore, studies on phosphorus removal and the recovery of phosphorus resources are important. In 2023, 27.3% of China's important fishery waters in inland rivers exceeded the total phosphorus standard. The area of marine natural important fishery waters that exceeded the standard for reactive phosphate was 27.0%, and the area of seawater key aquaculture areas that exceeded the standard for reactive phosphate was 28.2%. Aquaculture development is particularly important in the context of the 'Big Food Concept'. The China's total aquatic product output in 2023 was 71.16 million tons, an increase of 4.39% year-on-year, of which aquaculture production accounted for 81.6%. From 2022 to 2024, China's provinces and municipalities introduced the aquaculture tail water discharge standard. For example, Shandong Province has implemented DB37 4676-2023, which sets a total phosphorus primary discharge limit of 0.7 mg/L and a secondary discharge limit of 1.0 mg/L. Recently, the rapid development of aquaculture tailwater phosphorus removal technology and phosphorus recovery technology based on physical, chemical, biological and ecological methods has provided strong support for aquaculture tailwater phosphorus removal and recycling. The current aquaculture tailwater phosphorus removal technology has made some progress. However, the advanced removal of phosphorus from the tailwater and phosphorus recovery technology requires further investigation. Enhancing the advanced removal of aquaculture tailwater is essential to ensure the sustainable development of aquaculture. This study classified the phosphorus in the water, examined the principle and current status of aquaculture tailwater phosphorus removal technology, and reviewed the application of phosphorus removal in the tailwater of the current aquaculture model. The principles and current status of phosphorus removal technology in aquaculture tailwater were discussed in terms of physical, chemical, and biological methods of phosphorus removal. The study indicated that the physical method of phosphorus removal technology in aquaculture primarily relies on adsorption and membrane separation technology, in which the high

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adsorption saturation of adsorbent materials and renewable is the key to adsorption of phosphorus removal, and the physical principle of membrane separation technology is the selective permeability of the membrane. Pollutant retention is achieved through the concentration difference between the two sides of the membrane, hydraulic pressure difference, and potential difference. The current membrane separation technology research methods continue to innovate and generally combine the membrane separation technology and biological method applied to aquaculture tail water phosphorus removal technology. Chemical phosphorus removal technologies such as precipitation, electro-flocculation, crystallization and depth oxidation are important for aquaculture tailwater phosphorus removal technology. These technologies are notable in phosphorus resource recovery and should not be ignored. Biological phosphorus removal technology is a primary method for phosphorus removal in aquaculture tailwater and mainly includes biofilm reactor, Biofloc, microalgae biological purification, bacterial and algal synergistic reactor, artificial wetland and other technologies. Biofilm reactors and Biofloc mainly rely on the role of phosphate accumulating organisms (PAOs) and denitrifying phosphate accumulating organisms (DPAOs), both of which have different processing capacities and biological responses to phosphorus in aerobic, anaerobic and anoxic stages. PAOs absorb phosphorus in aerobic conditions and release phosphorus in anaerobic conditions; DPAOs release phosphorus in anaerobic conditions and absorb phosphorus in the anoxic stage. Microalgae biological purification technology mainly uses the photosynthesis of microalgae and microalgae growth to absorb and remove phosphorus from the water. The microalgae bioreactor is a bacterial-algae synergistic reactor formed by combining microalgae and biofilm reactors to remove phosphorus. Artificial wetlands are a comprehensive phosphorus management method that integrates physical, chemical, and biological methods. This approach is becoming prominent as a crucial technique for phosphorus management in aquaculture tailwater. Current aquaculture modes such as recirculating aquaculture system (RAS), pond aquaculture and other modes, in which RAS mostly use biofilm reactors, bacterial and algal synergistic bioreactors and multi-level integrated aquaculture systems and other treatment methods, and in recent years, artificial wetlands are also gradually applied in the treatment of phosphorus in RAS tailwater. Artificial wetlands are used with sediment and microbial fuel cells to remove phosphorus from aquaculture tailwater. In phosphorus treatment in recirculating aquaculture tailwater, the bioecological method is gradually being used as the main method to treat phosphorus in tailwater, supplemented by physicochemical methods. The pond aquaculture tailwater phosphorus management is also based on bioecological methods, such as 'three ponds and two dams', artificial wetlands, multi-level integrated aquaculture treatment system and other methods to remove phosphorus. This study analyzed the aquaculture tailwater phosphorus removal technology, which can provide new ideas for tailwater phosphorus treatment and phosphorus resource recovery and promote the green development of aquaculture.

Key words Phosphorus; Existing form; Aquaculture wastewater; Recirculating aquaculture; Phosphorus removal methods