



厌氧氨氧化及其处理低碳氮比氨氮废水的研究进展

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[摘要] 厌氧氨氧化是近 20 a 来逐步发展起来的一种高效、节能新型生物脱氮工艺, 其无需消耗碳源且污泥产量低, 是处理低碳氮比氨氮废水的较佳选择。对厌氧氨氧化工艺的基本原理、影响厌氧氨氧化细菌活性的常见因素作了简要介绍, 阐述了厌氧氨氧化工艺用于污水处理的发展历程, 引用部分案例对厌氧氨氧化工艺在低碳氮比废水处理中的应用状况进行了说明, 指出其对于低碳氮比高浓度氨氮废水处理效果较好, 但对于低碳氮比的中低浓度氨氮废水的处理存在技术瓶颈, 即中低浓度氨氮废水难以实现稳定的亚硝化。在此基础上对于解决中低浓度氨氮废水厌氧氨氧化处理瓶颈所取得的技术突破进行总结, 并展望了厌氧氨氧化技术在低碳氮比废水处理工程中的应用前景。

[关键词] 生物脱氮; 低碳氮比; 厌氧氨氧化; 亚硝化

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Anaerobic ammonia oxidation and its research progress for the treatment of low C/N ratio ammonia nitrogen wastewater

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Abstract: Anaerobic ammonia oxidation is a new type of biological nitrogen removal process with high efficiency and energy saving, which has been gradually developed in recent 20 years. Anaerobic ammonia oxidation does not need to consume carbon sources and has low sludge output, so it is the better choice for treating low C/N ratio ammonia nitrogen wastewater. This paper briefly introduced the basic principle of anaerobic ammonia oxidation process and common factors affecting the activity of AnAOB, described the development history of anaerobic ammonia oxidation process for sewage treatment. Also, it expounded the application of anaerobic ammonia oxidation process in the treatment of low C/N ratio wastewater by citing some cases, and pointed out its good effect on the treatment of low C/N ratio and high concentration ammonia nitrogen wastewater. However, there was a technical bottleneck in the treatment of low carbon nitrogen ratio and middle or low concentrated ammonia nitrogen wastewater, that was, it was difficult to achieve stable nitrosation of middle or low concentrated ammonia nitrogen wastewater. On this basis, the technical breakthroughs made in solving the bottleneck of anaerobic ammonia oxidation treatment of middle or low concentrated ammonia nitrogen wastewater were summarized, and the application prospect of anaerobic ammonia oxidation technology in the engineering of low carbon nitrogen ratio wastewater treatment was prospected.

Key words: biological nitrogen removal; low C/N ratio; anaerobic ammonium oxidation; nitrification

氨氮是废水中的主要污染物之一, 其过量排放 不仅会导致水体富营养化, 毒害水生生物, 也会对人

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体健康造成严重威胁^[1-2]。据2020年中国生态环境统计年报数据显示,全国氨氮全年排放量高达98.4万t,其中生活污水源废水、农业源废水和工业源废水中氨氮排放量分别占氨氮总排放量的71.85%、25.81%和2.13%^[3]。大部分氨氮废水,如城镇生活污水^[4]、垃圾渗滤液^[5]、养猪废水^[6-7]、稀土矿山废水^[8]等都存在碳源不足、碳氮比低的问题。低碳氮比废水的传统物理化学处理方法包括吹脱法^[9-10]、磷酸钙镁沉淀法(MAP法)^[11]、吸附法^[12-13]、折点加氯法^[14-15]、高级氧化法^[16-17]等。这些方法在去除氨氮的同时也存在诸多问题,如药剂消耗大、运行费用高、操作复杂、易引发二次污染等,因此限制了它们的工程化应用^[18-19]。生物脱氮技术包括硝化-反硝化工艺、亚硝化-反硝化工艺和同步硝化反硝化工艺等^[20-21]。相比于传统物理化学法,生物脱氮法因经济高效、环境友好的特点而得到更广泛应用。

硝化-反硝化脱氮工艺是目前应用最为广泛的生物脱氮技术,如目前污水处理厂大规模使用的AO法、A²O法、氧化沟法等工艺都是基于硝化-反硝化的工艺原理来进行废水中氨氮的脱除。反硝化过程需要消耗大量的碳源,理论上硝化-反硝化工艺脱除1g氮需要消耗约2.8g的碳源(以COD计),实际工程上碳源的消耗量要远远大于理论值,其 $m(\text{COD})/m(\text{NH}_3\text{-N})$ (记作C/N)往往高达5~6以上。对于碳源比较丰富、C/N大于6的污水,采用传统的硝化-反硝化工艺就能取得较好的氨氮和TN脱除效果。但对于C/N较低的废水,特别是C/N小于4的废水,若仍采用传统的硝化-反硝化工艺,则需要补充大量的碳源,相应地,处理过程中排放的二氧化碳及污泥产量也大幅增加,这些都增加了整体废水处理成本。此外,随着《合成氨工业水污染物排放标准》、《城镇污水处理厂污染物排放标准》等新的废水排放标准的实施,对于污水中TN的排放要求被进一步强化,寻找低碳降耗的新氨氮处理方法越来越重要。

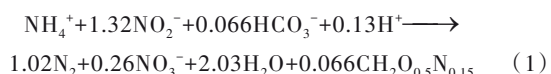
厌氧氨氧化(Anaerobic ammonium oxidation, ANAMMOX)自1995年首次被发现以来,因其节能高效、低碳、污泥产量低的特点,已逐渐发展成为最受欢迎的生物脱氮工艺之一^[22-23]。相比于硝化-反硝化工艺,ANAMMOX工艺可节省50%~60%的曝气能耗和100%的有机碳源^[24]。结合我国在2020年提出的“双碳”目标,ANAMMOX工艺应是处理低碳氮比氨氮废水的最优工艺选择之一。

笔者对ANAMMOX工艺的基本原理作了简要介

绍,回顾了ANAMMOX的发展历程,总结了ANAMMOX在低碳氮比废水处理中的研究现状和应用进展,并展望了ANAMMOX技术在我国低碳氮比废水处理中的广阔应用前景,以期ANAMMOX技术进一步的推广应用提供参考。

1 ANAMMOX工艺概述

ANAMMOX的主要原理是在厌氧或缺氧条件下,厌氧氨氧化细菌(AnAOB)以氨氮为电子供体,亚硝氮为电子受体,将氨氮氧化为 N_2 和少量硝态氮,其反应如式(1)所示。在这一过程中,涉及多种中间产物和功能酶。中间产物包括一氧化氮(NO)、肼(N_2H_4)等,功能酶包括亚硝酸还原酶(NIR)、肼合酶(HZS)、肼脱氢酶(HDH)^[25-26]等。



目前已知的AnAOB都归属于浮霉菌门(Planctomycetes)下的Brocadiales纲,主要有6个菌属,分别为*Candidatus_Brocadia*、*Candidatus_Kuenenia*、*Candidatus_Jettenia*、*Candidatus_Scalindua*、*Candidatus_Anammoxoglobus*、*Candidatus_Anammoximicrobium*^[27]。借助现代分子生物学技术,研究人员发现AnAOB体内有一个独特的膜结合隔室(Anammoxosome),其体积占据AnAOB总体积的50%~80%,在这里可以进行能量交换和特异性ANAMMOX反应^[28]。此外,AnAOB体内还含有大量自身产生的细胞色素,外观上呈鲜艳的红色。AnAOB繁殖速度慢,倍增周期为9~14d左右。有研究指出,AnAOB适宜的生长温度和pH分别为30~40℃和7.0~8.8^[29]。当温度高于45℃时,AnAOB的活性被严重抑制,且这种抑制不可逆,而低温条件(<20℃)对AnAOB的抑制在升温之后即可恢复^[24]。高浓度游离氨(FA)和游离亚硝酸(FNA)会抑制AnAOB的活性,M. TOMASZEWSKI等^[30]通过研究表明,FA、FNA分别低于20、0.01mg/L时有利于厌氧氨氧化工艺的脱氮。如式(2)、式(3)所示,pH会影响系统的FA与FNA浓度,pH偏低时,FNA浓度随之增加,pH偏高时,FA浓度会被提高,均不利于ANAMMOX工艺的脱氮^[31]。此外,Jinjin YU等^[32]还发现,过高或过低的pH也会对微量营养元素的有效性产生不利影响,进而影响系统的脱氮效果。

$$\text{FA} = \frac{17}{14} \times \frac{[\text{NH}_4^+ - \text{N}] \times 10^{\text{pH}}}{\exp\left(\frac{6334}{273 + T}\right) + 10^{\text{pH}}} \quad (2)$$

$$FNA = \frac{46}{14} \times \frac{[\text{NO}_2^- - \text{N}]}{\exp\left(\frac{2300}{273+T}\right) \times 10^{\text{pH}}} \quad (3)$$

AnAOB对环境变化较为敏感,在应用ANAMMOX工艺处理低碳氮比氨氮废水时,除上述提到的温度、pH、FA与FNA外,有机物、溶解氧(DO)、盐分等也都会影响ANAMMOX的脱氮性能。

AnAOB以无机碳为唯一碳源,废水中的有机物会对AnAOB的脱氮性能产生不同程度的影响,其主要表现在:(1)有机物的存在易引起反硝化细菌的滋生,其快速的生长使得AnAOB无法竞争获得足够的生长要素,进而削弱AnAOB的脱氮性能^[33];(2)有机物与氨氮竞争成为ANAMMOX反应的电子供体,以此演化出不同于ANAMMOX的代谢途径,从而减少氨氮的去除量^[34]。不少废水虽然C/N不够高,如C/N小于4,但因废水中存在有机物仍有可能影响ANAMMOX工艺,故要通过前置处理工序,将废水中的有机物尽量脱除,以保证ANAMMOX工艺的平稳运行。目前,通常采用前置反硝化工艺脱除原水中有有机物,脱除目标为 $m(\text{BOD}_5)/m(\text{NH}_3\text{-N}) < 1$,最好 $m(\text{BOD}_5)/m(\text{NH}_3\text{-N}) < 0.5$,然后再进入ANAMMOX处理单元。AnAOB对废水中FA和FNA有着不同的耐受程度,研究指出,当FA和FNA分

别大于各自的某个临界值时会对AnAOB的活性产生明显的抑制^[35]。研究表明,除先前提到的pH外,废水的温度、底物浓度等参数都会影响FA和FNA的浓度,进而影响ANAMMOX的脱氮效果^[36]。ANAMMOX反应需要在厌氧或者缺氧条件下进行,如果水中的DO高于18%氧饱和时的氧分压就会抑制AnAOB的活性,通过降低DO可恢复其活性^[37-38]。此外,高浓度的盐分会使得胞内外形成高渗透压,进而导致AnAOB质壁分离、休眠甚至死亡^[39]。

2 ANAMMOX工艺处理低碳氮比氨氮废水

ANAMMOX工艺包括基于亚硝化-ANAMMOX的SHARON-ANAMMOX工艺、OLAND工艺、CANON工艺和DEMON工艺等,各工艺优缺点如表1所示。付昆明等^[40]在对连续流CANON反应器运行稳定性进行研究时,在pH为7.39~8.01,温度为35℃条件下,得到系统平均TN去除负荷为1.8 kg/(m³·d);Rui DU等^[41]在上流式厌氧床反应器中采用一种新的气体混合方式运行连续流DEMON工艺,水力停留时间为0.5 h时,系统的最大TN去除负荷可达2.42 kg/(m³·d)。

表1 不同ANAMMOX工艺优缺点分析

Table 1 The advantage and disadvantage analysis of different ANAMMOX processes

工艺类型	优点	缺点
两段式厌氧氨氧化自养脱氮 SHARON-ANAMMOX	独立调节,灵活稳定;可避免有毒物质和有机物抑制厌氧氨氧化细菌;适合低浓度氨氮废水处理	占地面积大、投资成本高;易产生亚硝酸盐抑制
一体式厌氧氨氧化自养脱氮 OLAND CANON DEMON	占地面积小、投资成本低;适合处理高浓度氨氮废水,容积负荷大;可有效避免亚硝酸盐的抑制作用	需严格控制运行参数确保多种微生物菌群共存;启动时间长

世界范围内目前已有超过200座的ANAMMOX工程投入使用。国外在21世纪初就开展了ANAMMOX的工程化应用,2002年,荷兰鹿特丹Dokhaven市政污水处理厂构建了两段式亚硝化-ANAMMOX的SHARON-ANAMMOX工艺用于处理污泥消化液,历时3.5 a成功启动反应器,从而建立了世界上第一座ANAMMOX工程化装置^[42]。奥地利Strass污水处理厂在2003年采用DEMON工艺处理污泥脱水液,最高氮去除负荷达到0.7 kg/(m³·d)^[43]。之后,荷兰Olburgen工业污水处理厂采用一体式亚硝化ANAMMOX的CANON工艺处理污泥脱水液^[44],瑞典Sjölunda污水处理厂采用ANITATM-Mox工艺处理污泥消化液^[45],新加坡、美国等也相继建立了自己的第一座ANAMMOX

反应器。2009年,我国引进荷兰帕克公司的ANAMMOX技术用于处理味精生产废水^[46]。在后续的十余年间,我国在多种废水处理领域建设了ANAMMOX工程,如山东省滨州市安琪酵母公司处理酵母生产废水的ANAMMOX工程、北京市市政污水处理的污泥厌氧发酵液工程等。2021年,笔者所在研究团队在广东某印染纺织厂成功建设运行了世界上第一座处理印染丝光废水的ANAMMOX工程,该废水进水氨氮在1 500~5 000 mg/L波动,几乎不含有有机碳源,故可以直接使用ANAMMOX处理工艺进行处理,在不投加任何有机碳源条件下氨氮去除率大于95%,TN去除率大于85%,氨氮处理的容积负荷也达到1 kg/(m³·d)(以N计)。此外,本研究团队在2021年针对高铁列车集便器废水

和垃圾填埋场老龄垃圾渗滤液,也采用ANAMMOX工艺建设运行了高氨氮废水处理示范工程项目。虽然此类废水所含氨氮为500~2 500 mg/L,但其COD为1 000~6 000 mg/L,高浓度COD必然对ANAMMOX的工艺产生重大影响,因此在设计上采用了前置亚硝化-反硝化工艺,以消耗进水中存在的有机物。系统运行中,通过亚硝化反应器,将废水中的氨氮转化为亚硝酸根,之后经处理后的废水回流到前置亚硝化-反硝化反应器中,经过反硝化脱除大部分有机物,再进入ANAMMOX反应器,在低C/N条件下脱除氨氮。采用亚硝化-反硝化工艺的缺点是亚硝化的量不用严格控制,多余的亚硝酸根可作为后续ANAMMOX的反应基质。

图1简单示意了ANAMMOX工艺用于污水处理的发展历程,可以看出,ANAMMOX工艺从发现到真正大规模应用用了近7 a时间,成功解决了高氨氮废水的ANAMMOX工程化应用难题。

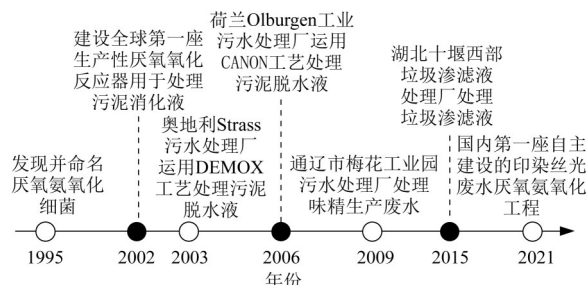


图1 ANAMMOX工艺用于污水处理的发展历程

Fig. 1 Development history of ANAMMOX process for sewage treatment

上述提及的ANAMMOX工程案例多为处理氨氮大于300 mg/L的高浓度氨氮废水。在“双碳”目标的驱动下,随着我国研究开发能力的不断增强,采用国内自行开发的ANAMMOX工艺处理低碳氮比高浓度氨氮废水的项目将会越来越多。

3 ANAMMOX对于低碳氮比的中低浓度氨氮废水处理的应用瓶颈及突破

采用低碳节能的ANAMMOX工艺处理高浓度的氨氮废水已进入大规模工业化阶段,但用其处理中低浓度的氨氮废水仍没有大规模工程化应用。中低浓度氨氮废水水量大,相较于高浓度氨氮废水,其排放的氨氮污染物总量更高。如何将ANAMMOX工艺成功应用到低碳氮比的中低浓度氨氮废水处理中,是目前水处理领域的研究热点之一。

由式(1)可知,ANAMMOX的反应底物主要为

氨氮和亚硝氮,其物质的量比为1.32:1。因此在应用ANAMMOX工艺处理氨氮废水时,需要将部分氨氮转化为亚硝氮。稳定的亚硝化是ANAMMOX进行的前提,也是中低浓度氨氮废水实现ANAMMOX处理的技术瓶颈。

高浓度氨氮废水中氮亚硝化技术已经趋于成熟,亚硝化策略包括高温^[47],低DO^[48],低污泥龄(SRT)^[49],对曝气量、碱度等参数的实时控制^[50],对FA和FNA的控制等^[51]。罗远玲等^[52]采用序批式反应器(SBR),以具有良好亚硝化积累的颗粒污泥为接种污泥,考察了温度变化对系统氮转换性能的影响,结果表明,相较于15、25℃,温度为30℃时,系统具有良好的短程硝化性能,出水亚硝氮积累率为93.17%。Yongyuan YANG等^[53]利用沸石的吸附-再生性能,对污水中的氨进行有效吸附及脱附,将水中FA保持在适当的浓度范围内,实现了氮素良好的亚硝化。

对于中低浓度氨氮的ANAMMOX,必须要有稳定的亚硝态氮来源来保障ANAMMOX工艺的稳定运行。短程反硝化工艺可利用小部分有机碳源将硝态氮还原成亚硝态氮而不进一步还原成N₂,以此为中低浓度氨氮废水的ANAMMOX处理提供亚硝态氮^[4,41]。Lingxiao GONG等^[54]利用饥饿法在SBR反应器中成功实现短程反硝化,NO₃⁻-N→NO₂⁻-N转化率(NTR)达71.7%。Shenbin CAO等^[55]在USB反应器中安装了一套气体自动循环装置,减少了反应器死区,从而实现了高达81.4%的NTR。Rui DU等^[56]采用DEAMOX工艺,构建了以醋酸盐和乙醇为有机碳源的2个反应器用于处理含氨氮、硝态氮均为50 mg/L的模拟废水,其NTR分别达到95.8%和87.0%。此外该团队采用短程反硝化-厌氧氨氧化工艺(PD-A)处理北京工业大学校园内氨氮为57.87 mg/L的生活污水,出水TN可稳定低于5 mg/L^[57]。研究表明,碳源种类、碳氮比、温度、pH等参数均会影响短程反硝化效果^[58-60],实际应用中均需加以控制^[58-60]。

另外,探索中低浓度氨氮的稳定亚硝化,是另外一条解决中低浓度氨氮废水ANAMMOX处理时亚硝态氮来源短缺问题的途径。Zhenguo CHEN等^[61]研究了采用沸石生物固定床(ZBFB)反应器处理低浓度氨氮废水(50 mg/L)的亚硝化性能,结果表明,当床层温度升至36℃时,系统达到稳定亚硝化效果。对于中低浓度氨氮废水,有研究通过投加少量的双氧水,利用双氧水抑制NOB能力强、抑制AOB能力弱的特性,达到中

低浓度氨氮废水的稳定亚硝化^[62-63]。

城市污水处理厂的主流 ANAMMOX 工艺,因市场巨大,TN 负荷削减明显,引起了众多研究者的关注。但是由于城市污水处理厂进水氨氮只有 20~50 mg/L,其亚硝氮的获取更加困难,导致目前多数研究都限于实验室小试或中试,鲜有工程化应用的报道。2018 年通过运行数据分析,研究人员发现西安第四污水处理厂出水的 TN 只有 5 mg/L,经进一步的分析,确认处理系统中存在部分 ANAMMOX。这一重大发现,表明了 ANAMMOX 应用于主流污水处理系统具有可行性。但市政污水的氨氮浓度低、水量巨大,要实现 ANAMMOX,研究人员必须克服和解决以下几个问题:(1)氨氮与经转化形成的亚硝酸根浓度都很低,对亚硝酸氧化菌不可能形成抑制,形成的亚硝酸根很容易在水中少量溶解氧存在的条件下被氧化而形成硝酸根;(2)厌氧氨氧化菌本身增殖速度很慢,在低浓度氨氮条件下,增殖速度更慢,且在污水处理高流量的条件下,很容易流失;(3)城市污水的水温及其他水质条件存在波动,影响 ANAMMOX 工艺的稳定性。

4 总结与展望

ANAMMOX 工艺无需投加碳源、曝气量少、能耗低、污泥产量低,是处理低碳氮比氨氮废水的最佳选择。高浓度氨氮废水的 ANAMMOX 工艺已经成熟,在“双碳”目标的驱动下将会得到更加广泛的应用。但在中低浓度氨氮废水处理领域,尤其是在市政污水处理领域,由于进水基质浓度低、温度随四季变化大,亚硝氮的稳定获取往往比处理高浓度氨氮废水时困难得多,采用 ANAMMOX 对其进行处理的工程案例鲜有报道。近年来,研究者们积极探索基于短程反硝化或稳定亚硝化的边界条件,已取得了较多的研究成果,但因在实际工程应用中该技术仍存在诸多不确定影响因素,导致其大多仍停留在实验室阶段,并未真正大规模工程化应用。因此,中低浓度氨氮废水的 ANAMMOX 研究,市政污水的主流 ANAMMOX 研究及工程化应用开发将是今后的重点研究目标。

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