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磷酸盐对亚硝化系统的抑制及恢复

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摘要: 通过接种城镇污水处理厂的污泥, 采用连续流反应器启动亚硝化系统并改变进水磷酸盐的浓度, 研究了不同磷酸盐浓度对亚硝化系统的影响。结果表明经过 14 d 的运行, 亚硝化系统启动成功, 氨氮转化率达到 92.2%, 亚硝酸盐累积率为 73.66%, 亚硝酸盐产生速率达到 $14.42 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$ 。磷酸盐浓度在 $10 \sim 30 \text{ mg} \cdot \text{L}^{-1}$ 时对亚硝化系统的影响并不大; 随着磷酸盐浓度持续提高, 氨氮转化率在不断降低。当磷酸盐的浓度为 $80 \text{ mg} \cdot \text{L}^{-1}$ 时, 系统的氨氮转化率为 13.6%, 亚硝酸盐累积率仅 18.19%, 亚硝酸盐产生速率仅 $0.54 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$, 亚硝化反应受到严重抑制。将进水磷酸盐浓度降低到 0, 经过 14 d 运行, 亚硝化系统获得恢复, 且氨氮转化率可以达到 80% 以上, 亚硝酸盐累积率达到 86.96%, 亚硝酸盐产生速率为 $15.63 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$ 。

关键词: 亚硝化系统; 连续流; 磷酸盐; 抑制; 恢复

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Inhibitory Effects of Phosphate and Recovery on a Nitrification System

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Abstract: The effect of phosphate concentration on nitrification was studied by using a stabilization nitrosation system, which was started up in a continuous flow reactor by inoculating sludge from a municipal wastewater treatment plant. The results showed that the nitrification system was started successfully after operating for 14 days. The conversion rate of ammonia nitrogen reached 92.2%, the nitrite accumulation rate was 73.66%, and the nitrite generation rate was $14.42 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$. There was no effect of phosphate concentration on the nitrosation system between 10 and $30 \text{ mg} \cdot \text{L}^{-1}$; and the conversion rate of ammonia nitrogen was decreased with the continuous increase in phosphate concentration. When the concentration of phosphate was $80 \text{ mg} \cdot \text{L}^{-1}$, with an ammonia conversion rate 13.6%, accumulation rate of nitrite of 18.19%, and nitrite generation rate of $0.54 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$, the reaction was severely inhibited. After reducing the influent phosphate concentration to 0, with the ammonia nitrogen conversion rate at more than 80%, nitrite accumulation rate improved to 86.96%, and the nitrite generation rate being $15.63 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$, the system recovered after operating for 14 days.

Key words: nitrosation system; continuous flow; phosphate; inhibition; recovery

厌氧氨氧化是指在厌氧条件下, 一类微生物能够利用氨氮作为电子供体, 亚硝酸盐作为电子受体, 通过生物氧化还原作用转化为氮气的生化过程。因厌氧氨氧化脱氮过程具有无需添加有机碳源, 脱氮速率高, 污泥产量低等^[1-5]优点而受到研究者的关注。然而厌氧氨氧化需要亚硝酸盐作为电子受体, 而一般的工业废水中仅含有氨氮, 因此厌氧氨氧化需要与亚硝化联合, 才能实现废水中氨氮去除。亚硝化作为厌氧氨氧化的前置步骤, 决定着厌氧氨氧化脱氮效能的高低。

近年来对影响亚硝酸盐积累因素如 DO、pH、温度、COD/N、接种污泥等的研究较多。目前认为亚硝化反应的最适 pH 范围为 7.5 ~ 8.0^[6-10], 最适的温度在 $30 \sim 37^\circ\text{C}$ ^[11-13], DO 浓度不高于 $1 \text{ mg} \cdot \text{L}^{-1}$ ^[14-19], 一般控制在较低水平有利于 AOB 的生长。

然而一些工业废水中除了氨氮, 还有重金属、有机物、磷酸盐等一些污染物。这些物质对部分亚硝化-厌氧氨氧化系统会产生影响。虽然目前有研究者关注到磷酸盐对厌氧氨氧化的影响^[20-23], 然而亚硝化作为联合工艺的一部分, 却很少有研究者关注磷酸盐对其的影响。因此研究磷酸盐对亚硝化的影响对联合工艺工程化应用具有重要意义。本研究研究了不同磷酸盐的浓度对亚硝化系统的抑制强弱, 以期对厌氧氨氧化联合工艺在实际工程中的运行提供依据。

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1 材料与方法

1.1 反应器与运行参数

本实验采用 $15\text{ cm} \times 15\text{ cm} \times 40\text{ cm}$ 的反应器,有效容积为 7 L ,内置膜状填料.接种苏州市城镇污水厂取样所得的污泥于反应器中.通过蠕动泵控制进水流量,控制水力停留时间(HRT) 1.3 d .反应器内部放有恒温加热棒来控制温度为 30°C ,反应器底部设有微孔曝气头,由气泵供气,通过转子流量计控制气量,控制溶解氧为 $0.5\text{ mg}\cdot\text{L}^{-1}$.通过投加药剂控制 pH 在 $7.8 \sim 7.9$ 之间.

1.2 进水水质

本实验采用人工模拟废水.废水的主要组成: $1\ 128\text{ mg}\cdot\text{L}^{-1}\text{ NH}_4\text{HCO}_3$, NaHCO_3 (调节 pH 和提供碳源), KH_2PO_4 及微量元素浓缩液 I ($1\text{ mL}\cdot\text{L}^{-1}$): $5\ 000\text{ mg}\cdot\text{L}^{-1}\text{ EDTA}$, $5\ 000\text{ mg}\cdot\text{L}^{-1}\text{ FeSO}_4$; 微量元素浓缩液 II ($1.25\text{ mL}\cdot\text{L}^{-1}$): $5\ 000\text{ mg}\cdot\text{L}^{-1}\text{ EDTA}$, $430\text{ mg}\cdot\text{L}^{-1}\text{ ZnSO}_4\cdot 7\text{ H}_2\text{O}$, $250\text{ mg}\cdot\text{L}^{-1}\text{ CuSO}_4\cdot 5\text{ H}_2\text{O}$, $240\text{ mg}\cdot\text{L}^{-1}\text{ CoCl}_2\cdot 6\text{ H}_2\text{O}$, $990\text{ mg}\cdot\text{L}^{-1}\text{ MnCl}_2\cdot 4\text{ H}_2\text{O}$, $220\text{ mg}\cdot\text{L}^{-1}\text{ NaMoO}_4\cdot 2\text{ H}_2\text{O}$, $190\text{ mg}\cdot\text{L}^{-1}\text{ NiCl}_2\cdot 6\text{ H}_2\text{O}$, $210\text{ mg}\cdot\text{L}^{-1}\text{ NaSeO}_4\cdot \text{H}_2\text{O}$, $14\text{ mg}\cdot\text{L}^{-1}\text{ H}_3\text{BO}_3$.

1.3 分析方法

各种指标检测方法参照文献[24]. $\text{NH}_4^+\text{-N}$: 纳氏试剂分光光度法; $\text{NO}_2^-\text{-N}$: N (1-萘基)-乙二胺分光光度法; $\text{NO}_3^-\text{-N}$ 和 $\text{PO}_4^{3-}\text{-P}$ 采用离子色谱(戴安 IC-900, 美国)测定; pH 值采用 pH-3E 型酸度计测定; DO 采用溶氧仪测定.

2 结果与讨论

2.1 亚硝化系统的启动

反应器运行 5 d 后,测得系统中的 pH 为 7.51 ,出水氨氮和硝酸盐浓度呈上升趋势(图 1),而亚硝酸盐浓度呈下降趋势.谢新力等[26]的研究表明:适宜硝化细菌的 pH 为 $7.5 \sim 8.0$,适宜亚硝化细菌的 pH 为 $7.8 \sim 7.9$.薛源等[25]在探究亚硝化工艺的影响因素中发现 pH 值对亚硝化工艺存在影响,且亚硝化系统运行的 pH 应控制在 $7.8 \sim 7.9$.pH 过低会降低亚硝化反应的效率,抑制系统的氨氮转化,导致出水的氨氮浓度增加.当系统的 pH 为 7.51 时,硝化细菌将系统积累的亚硝酸盐进一步氧化成硝酸盐,进而出水的硝酸盐浓度呈增长趋势.因此,用碳酸氢钠调节碱度使部分亚硝化系统的 pH 到 7.85 .调节 pH 以后,出水氨氮浓度从 $64.37\text{ mg}\cdot\text{L}^{-1}$ 降低

到 $14.08\text{ mg}\cdot\text{L}^{-1}$,亚硝酸盐浓度从 $99.7\text{ mg}\cdot\text{L}^{-1}$ 上升到 $131.23\text{ mg}\cdot\text{L}^{-1}$,硝酸盐浓度上升幅度不大,由此可见 pH 对亚硝化反应的影响很大.

维持进水氨氮浓度在 $200\text{ mg}\cdot\text{L}^{-1}$ 左右,系统运行 14 d 后,出水氨氮浓度稳定在 $14.2\text{ mg}\cdot\text{L}^{-1}$ 、亚硝酸盐浓度稳定在 $130.15\text{ mg}\cdot\text{L}^{-1}$,硝酸盐浓度稳定在 $40\text{ mg}\cdot\text{L}^{-1}$,系统的 $\text{NH}_4^+\text{-N}$ 转化率达 92.2% 以上,说明亚硝化系统启动成功.

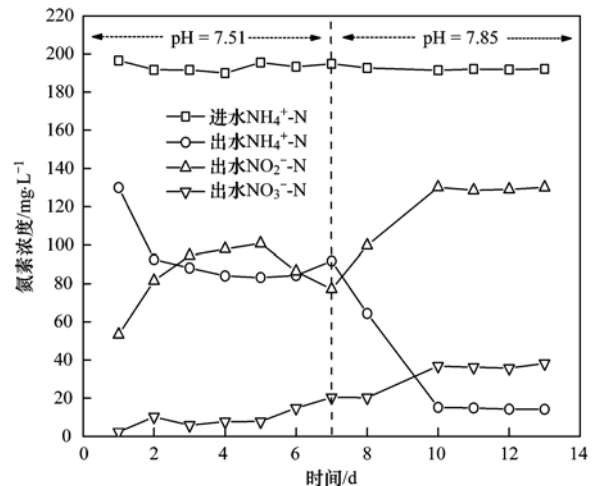


图 1 亚硝化系统的启动

Fig. 1 Start of the nitrification system

2.2 磷酸盐对亚硝化系统的影响

初始投加浓度为 $10\text{ mg}\cdot\text{L}^{-1}$,控制其它运行条件不变,一段时间后,出水水质如图 2 所示.当磷酸盐浓度为 $10\text{ mg}\cdot\text{L}^{-1}$ 时,亚硝化系统没有出现明显的抑制,系统氨氮转化率为 91.4% ,亚硝酸盐积累率为 68.86% ,亚硝酸盐产生率为 $13.33\text{ g}\cdot(\text{m}^3\cdot\text{d})^{-1}$.王鹏等[27]的研究认为在盐度微量的条件下亚硝化反应能正常进行而不受影响.王惠等[28]认为由于磷是微生物生长代谢不可或缺的基质,当少量的磷酸盐加入系统时,该部分磷酸盐主要作为系统微生物的生长基质而被微生物吸收利用,因此对亚硝化反应的影响微弱.由此可以判断进水磷酸盐浓度为 $10\text{ mg}\cdot\text{L}^{-1}$ 时不足以对多数的亚硝化污泥活性产生影响,亚硝化系统受磷酸盐的影响不大.

继续提高进水磷酸盐浓度为 $30\text{ mg}\cdot\text{L}^{-1}$,系统运行一段时间后,出水氨氮浓度呈增长趋势、出水亚硝酸盐浓度持续下降、出水硝酸盐浓度出现平稳增长,亚硝化反应受到了一定的抑制.

有研究表明,系统中加入了适宜浓度的磷酸盐后,其中少部分磷酸盐作为微生物生长所必要的生长基质而被微生物所吸收利用.而大部分的磷酸盐

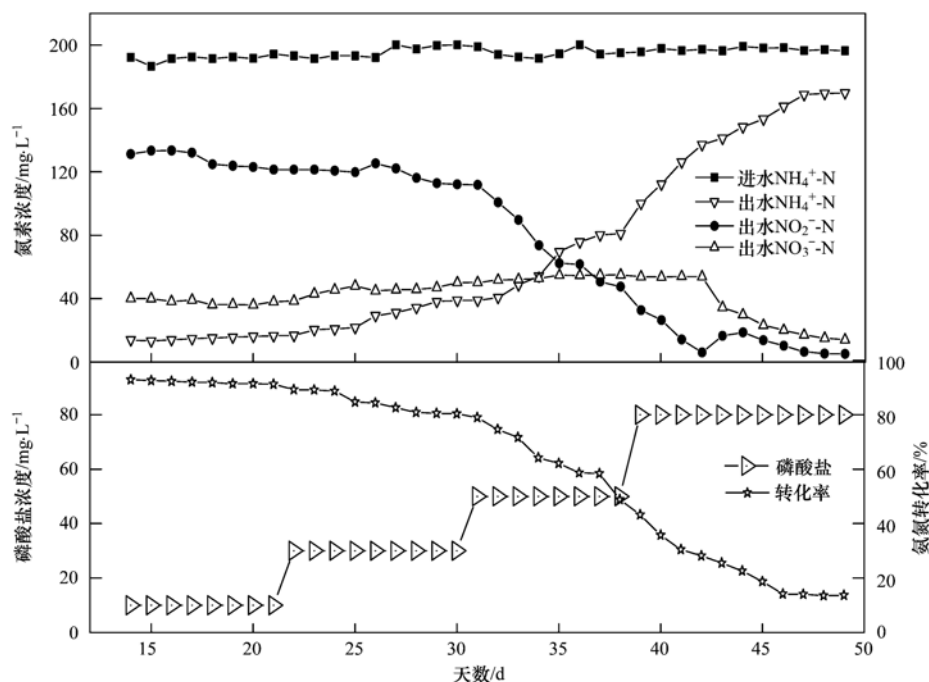


图2 磷酸盐对亚硝化系统的影响

Fig. 2 Effect of phosphate on nitrosation system

对亚硝化反应有抑制作用,很大程度地抑制了亚硝化菌的活性而使得系统氨氮转化率下降. 王鹏等^[27]的研究发现:进水盐度达一定浓度时,亚硝化系统会受到抑制,氨氮转化率降低. Daalkhaijav等^[2]的研究发现,一定浓度的磷酸盐对硝化反应有着相当程度的促进作用,由于系统中磷酸盐的存在促进了硝化反应的进行和发生. 这也能解释出水中硝酸盐浓度增长的现象. 因此造成出水亚硝氮浓度降低的原因可能是:①由于磷酸盐对亚硝化反应的抑制性作用,降低了系统的氨氮转化率,进而系统中亚硝酸盐的积累量也随之降低了;②系统中的硝化细菌将亚硝酸盐转化,导致出水亚硝酸盐的浓度降低,而硝酸盐浓度呈增长趋势.

当进水磷酸盐浓度进一步提高到 $50 \text{ mg} \cdot \text{L}^{-1}$ 后,出水氨氮浓度显著增加,出水亚硝酸盐浓度明显降低,出水硝酸盐浓度继续保持平稳增长. 系统运行到第 39 d 时,出水氨氮浓度为 $100.12 \text{ mg} \cdot \text{L}^{-1}$ 、亚硝酸盐浓度为 $32.76 \text{ mg} \cdot \text{L}^{-1}$ 、硝酸盐浓度为 $53.81 \text{ mg} \cdot \text{L}^{-1}$. 氨氮转化率为 48.8%,亚硝酸盐积累率为 34.3%,亚硝酸盐产生率为 $3.6 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$. 当磷酸盐浓度为 $80 \text{ mg} \cdot \text{L}^{-1}$ 时,运行一段时间后,系统中的氨氮转化率从 43.1% 降到了 13.6%,亚硝酸盐积累率降到 18.19%,亚硝酸盐产生速率只有 $0.54 \text{ g} \cdot (\text{m}^3 \cdot \text{d})^{-1}$. 说明浓度为 $80 \text{ mg} \cdot \text{L}^{-1}$ 的磷酸盐对亚硝化反应的抑制作用很强. 磷

酸盐对亚硝化系统产生抑制的机制为:一定浓度的盐分会对硝化细菌的细胞形态及菌群结构产生破坏,致其发生不可逆改变,由于亚硝化菌群失活从而降低系统的氨氮转化率. 当投入的磷酸盐浓度处于 $10 \sim 80 \text{ mg} \cdot \text{L}^{-1}$ 范围时,随磷酸盐浓度的提高,亚硝化系统受到的抑制作用也越来越大. 这一结论与 Geets 等^[29]的研究结果相似,他们的结果显示:磷酸盐浓度在 $40 \sim 100 \text{ mg} \cdot \text{L}^{-1}$ 时,亚硝化工艺出现明显的抑制现象. 而本实验结果是在 $30 \text{ mg} \cdot \text{L}^{-1}$ 时出现了明显的抑制现象.

运行到 50 d 后,测定出水氨氮浓度为 $170.45 \text{ mg} \cdot \text{L}^{-1}$ 、出水亚硝酸盐浓度为 $4.89 \text{ mg} \cdot \text{L}^{-1}$ 、出水硝酸盐浓度为 $14.01 \text{ mg} \cdot \text{L}^{-1}$. 此时,系统的氨氮转化率为 13.6%,氨氧化菌(AOB)处于完全抑制状态. 持续提高系统中磷酸盐浓度,系统出水氨氮浓度、亚硝酸盐浓度、硝酸盐浓度趋于稳定. 研究表明:磷酸盐抑制亚硝化系统的极限浓度是 $80 \text{ mg} \cdot \text{L}^{-1}$,此浓度下的系统氨氮转化率很低、亚硝酸盐积累量很小、AOB 的活性已完全抑制. 之后,随着磷酸盐浓度的增大,磷酸盐对系统的抑制作用趋于饱和,系统各出水浓度波动不大.

当磷酸盐浓度为 $10 \text{ mg} \cdot \text{L}^{-1}$ 时,系统的氨氮转化率为 91.2%. 随磷酸盐浓度的增加,系统的氨氮转化率逐渐下降,但不呈线性关系. 本研究发现:当加入系统的磷酸盐浓度微量时,磷酸盐对系统的氨

氮转化率抑制程度较小,从而出水氨氮浓度变化较小,微量的磷酸盐被系统中的微生物吸收利用,所以对亚硝化反应的影响很小;当流入系统的磷酸盐浓度 $30 \sim 80 \text{ mg} \cdot \text{L}^{-1}$ 时,系统中的氨氮转化率由 91.2% 变为 13%,呈现明显的下降趋势,同出水亚硝酸盐浓度相相应,说明了随着进水磷酸盐浓度的升高,亚硝化反应受到的抑制越来越显著,最后亚硝化反应趋于停止。

2.3 亚硝化污泥活性的短期恢复

停止向进水中加入磷酸盐,进水氨氮浓度为 $200 \text{ mg} \cdot \text{L}^{-1}$ 左右,控制反应器运行条件不变。系统运行一段时间后,部分亚硝化系统中各组分的浓度变化如图 3 所示。出水的氨氮浓度在持续降低最终达到稳定、出水亚硝酸盐浓度越来越高,最终达到稳定、出水硝酸盐浓度基本保持不变。

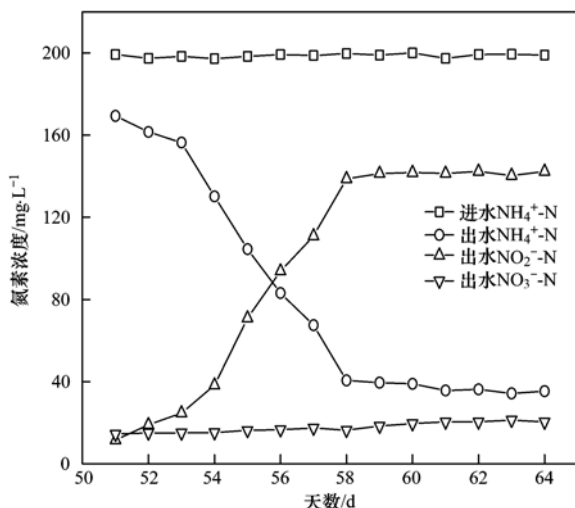


图 3 亚硝系统的恢复

Fig. 3 Recovery of nitrite system

亚硝酸盐浓度的变化趋势表明亚硝化系统在逐步恢复活性,控制系统合适运行条件如:温度、pH、DO 等可以使得亚硝化系统在短期内达到恢复。当系统运行的第 60 d 起,测定出水的氨氮浓度为 $38.92 \text{ mg} \cdot \text{L}^{-1}$ 、出水亚硝酸盐浓度为 $141.75 \text{ mg} \cdot \text{L}^{-1}$ 且之后的系统运行过程中波动不大,表明部分亚硝化系统得以恢复且已运行稳定,亚硝酸盐积累率达到 80% 以上。结果表明抑制后的 AOB 比硝化菌(NO_B)恢复得更快,导致恢复后亚硝酸盐的积累率比抑制前还高。孙洪伟等^[30]的研究得出游离铵(FA)对 AOB 活性的抑制作用是可逆的,而对 NO_B 活性的抑制作用是不可逆。说明 NO_B 抑制后恢复不了,使得系统中 AOB 成为优势菌体,亚硝酸盐可以得到更多的积累。

3 结论

(1)控制亚硝化系统高效运行的外界条件是:温度 30°C 、pH $7.8 \sim 7.9$ 、DO $0.5 \text{ mg} \cdot \text{L}^{-1}$,控制进水氨氮 $200 \text{ mg} \cdot \text{L}^{-1}$,运行 14 d,系统实现稳定运行,氨氮转化率达到 92.2%。

(2)一定浓度的磷酸盐对部分亚硝化工艺的运行有抑制性影响,当磷酸盐浓度在 $10 \sim 30 \text{ mg} \cdot \text{L}^{-1}$ 时,对 AOB 抑制作用不明显;随着磷酸盐浓度继续增大,磷酸盐对 AOB 的抑制效果越明显,且随磷酸盐浓度梯度的增加,抑制能力越来越强;当磷酸盐浓度 ($> 80 \text{ mg} \cdot \text{L}^{-1}$) 很大时,AOB 的活性被完全抑制了。

(3)通过停止磷酸盐的投加,稳定运行一段时间后,亚硝化系统污泥可以恢复活性,氨氮转化率可以达到 80% 以上。

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