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Effect of Reactor Stages in Series in the Main Anoxic Section on Anoxic Phosphorus Absorption Performance of Modified A^2O Process

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Abstract: Based on the kinetics of the treatment process of the completely mixed reactor in series, this study reveals the relationship between the reactor stages and the treatment efficiency, and it was applied to the simultaneous nitrogen and phosphorus removal process. The strengthening effect of the reactor stages of the main anoxic sections on the anoxic phosphorus absorption efficiency and the contribution to improving the treatment effect were investigated. Using sewage with a low carbon-to-nitrogen ratio as the research object and keeping the operation parameters of the improved anaerobic–anoxic–oxic (A^2O) process unchanged, the experimental research was carried out under the condition that reactor stages in series of the main anoxic section were one, two, three and four, respectively. The results showed an increase in the number of reactors in series in the main anoxic zone. The total phosphorus (TP) concentration in the effluent of the main anoxic stage decreased significantly, and the phosphorus uptake increased from 4.411 g/d (when n; the number of reactor stages in series was one) to 5.086 g/d when n was 4. Additionally, the nitrate nitrogen (NO_3^-–N) concentration in the effluent decreased, from 12.53 mg/L when n was one, to 9.62 mg/L when n was four, the removal rate of total nitrogen (TN) increased, from 56.86% when n was one to 65.98% when n was four, and the reduction power of nitrate nitrogen increased, and the denitrification rate increased. The increase in the number of reactors in series enhanced the anoxic phosphorus absorption and denitrification performance. Therefore, the main anoxic section of the synchronous nitrogen and phosphorus removal system can be designed and operated as reactors in series.

Keywords: hypoxia and phosphorus absorption; the number of reactor stages in series; denitrification; improved A^2O

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1. Introduction

Poor water environment quality and severe damage to water ecology have a substantial negative impact on people's health and are not conducive to sustainable economic and social development. To strengthen the prevention and control of water pollution and ensure water safety, experts have conducted many water treatment studies and achieved remarkable results. The overall steady improvement of water environment quality is due to the completion and operation of many sewage treatment plants [1–3]. The primary pollutants in sewage include organic matter, nitrogen and phosphorus, and other unique pollutants. Especially in industrial wastewater, the composition of pollutants is even more complex. According to different pollution components, sewage treatment methods can be divided into physical, chemical and biological treatment methods, including the adsorption method and catalytic oxidation method, with the help of environmental functional materials, which have been widely studied in recent years [4,5]. Among the methods mentioned above, most of the physical methods are used in the pretreatment part of the sewage treatment

process, and chemical, adsorption, catalytic oxidation and other methods are mainly used to treat smaller volumes of water, refractory or toxic wastewater. The treatment effect is good, but the treatment cost is high; the biochemical method is most widely used because of its low operation cost, especially in large-scale urban sewage treatment plants. Currently, the main core processes of urban sewage treatment plants are all biological treatment methods, and about 95% are activated sludge methods [6–10]. The reactor types include a horizontal flow reactor, a complete mixing reactor, a complete mixing reactor in series and a batch-activated sludge reactor. The flow pattern of sewage in the complete mixing reactor in series is between the complete mixing reactor and the horizontal flow reactor, which is an ideal hydraulic flow pattern. According to research reports, the use of a complete mixing reactor in series can increase the concentration gradient of pollutants in the reactor, improve the reaction efficiency, give full play to the degradation function of activated sludge microorganisms, increase the reaction rate of substances, and improve the treatment effect of sewage [11–18]. In addition, according to research reports, no matter in which field, when the treatment capacity and output is equal, the reactor's total volume or total reaction time is related to the number of series reactors [19,20].

With the increasing need to protect freshwater ecology, nitrogen and phosphorus are the main removal parameters of sewage treatment plants. Therefore, simultaneous nitrogen and phosphorus removal from sewage, promoting denitrification and phosphorus absorption, can reduce energy consumption, reduce excess sludge, and alleviate the contradiction between phosphorus-accumulating bacteria and denitrifying bacteria competing for carbon sources. This is especially suitable for treating sewage with a low carbon-to-nitrogen ratio [21–28]. In this paper, based on the kinetics of the treatment processes of a completely mixed reactor in series, the relationship between the reactor stages and the treatment efficiency is discussed, as it was applied to the simultaneous nitrogen and phosphorus removal process. Furthermore, the strengthening effect of the reactor stages in a series of the main anoxic sections on the anoxic phosphorus absorption efficiency and the contribution to improving the treatment effect was investigated. A feasible method for strengthening the application of the denitrifying phosphorus absorption process in a sewage treatment system is provided, which is of great significance for alleviating water environment pollution and water resource shortage.

2. Kinetics of Treatment Process in Series Complete Mixing Reactor

For the system with *n*-stage complete mixing reactors in series, the output stream of the previous complete mixing reactor serves as the input stream of the next stage reactor. Figure 1 shows the multi-stage series complete mixing reactor model.

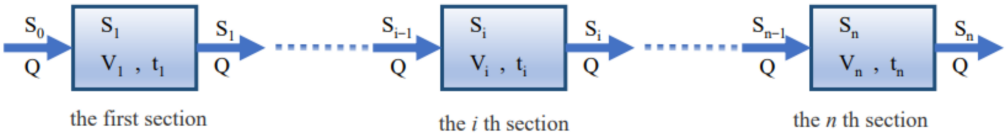

Figure 1. Model of a complete mixing reactor in series.

It can be seen in Figure 1 that the steady-state formula of the material balance in the *i*th reactor is as follows:

$$\frac{dC_i}{dt_i}V_i = 0 = QC_{i-1} - QC_i + r_{ci}V_i \quad (1)$$

Assuming that the removal of pollutants is the first-order reaction kinetics ($r_{ci} = -kC_i$), Formula (1) is arranged to obtain C*i* as

$$C_i = \frac{C_{i-1}}{1 + kt_i},\ i = 1,\ 2, \ldots,\ \mathrm{n} \quad (2)$$

where:

Q—influent water, m^3/h;

C_i—the mass concentration of pollutants in the ith reactor, g/m^3;

V_i—ith reactor volume, m^3;

r_{ci}—the reaction rate of substance C in the ith reactor, $g/(m^3 \cdot h)$;

t_i—hydraulic retention time (HRT) of the ith reactor, h;

k—degradation rate constant of pollutants, 1/h.

For multi-stage series-connected complete mixing reactors, under the condition of constant total residence time, when the HRT of each stage reactor is the same ($t_i = t/n$), the system can realize the optimal operation. Therefore, when the system operates under optimal conditions, the pollutant concentration C_n in the effluent of the nth reactor is

$$C_n = C_0/(1 + kt/n)^n \quad (3)$$

where:

C_o—influent pollutant mass concentration, g/m^3;

C_n—the mass concentration of pollutants in the nth reactor, g/m^3;

t—total hydraulic retention time, h.

From Formula (3), we can calculate the total hydraulic retention time and total effective volume of the system:

$$t = [(C_0/C_n)^{1/n} - 1]n/k \quad (4)$$

$$V = nQ/k[(C_0/C_n)^{1/n} - 1] \quad (5)$$

Table 1 elaborates on the relationship between different treatment efficiencies, different reactor stages in series, and the total reactor volume required. The volumes required for the horizontal flow reactors are also shown in the table for comparison purposes.

Table 1. Relationship between the number of reactor stages in series, the treatment efficiency, and the total required reactor volume.

The Number of Reactors in Series	Required Volume (in $V/(Q/k)$)			
	85% Removal Rate	90% Removal Rate	95% Removal Rate	98% Removal Rate
1	5.67	9.00	19.00	49.00
2	3.16	4.32	6.94	12.14
4	2.43	3.11	4.46	6.64
6	2.23	2.81	3.89	5.52
8	2.14	2.67	3.63	5.05
10	2.09	2.59	3.49	4.79
Horizontal flow	1.90	2.30	3.00	3.91

Note: the volume of a single reactor is equal to the value in the table divided by the number of reactors in a series.

3. Materials and Methods

3.1. Simulated Domestic Sewage

Simulated domestic sewage was set as the investigation object in the continuous flow test, and wastewater from a brewery was the source of chemical oxygen demand (COD). The concentration of ammonia nitrogen was prepared by adding NH_4Cl into the mixture; the influent phosphate concentration was prepared by adding KH_2PO_4. During the water distribution process, $NaHCO_3$ was added to supplement the alkalinity of the influent to meet the requirement of alkalinity in the nitrification reaction. The addition of $MgSO_4$ and $CaCl_2$ met the requirements of Mg^{2+} and Ca^{2+} plasma in the process of phosphate-accumulating bacteria. The addition of the trace element solution helped to meet the nutritional requirements of the growth and reproduction of the active sludge microorganism. According to different test requirements, the daily preparation of domestic sewage concentration maintained the constant influent COD concentration during the test. The NH_4Cl dosage was changed to adjust the influent ammonia nitrogen concentration,

and the KH_2PO_4 dosage changed to adjust the influent phosphorus concentration. See Table 2 for the simulated wastewater composition and water quality characteristics.

Table 2. Composition and water quality characteristics of test simulated wastewater.

Compound	Dosing Amount/ (g/L)	Composition of Trace Element Liquid	Concentration/ (g/L)	Water Quality Index	Concentration/ (mg/L)
Brewery wastewater	2~3 mL/L	$FeCl_3$	0.9	$CODc_r$	300 ± 10
NH_4Cl	0.1~0.24	H_3BO_4	0.15	biochemical oxygen demand (BOD_5)	172 ± 8
KH_2PO_4	0.031~0.035	$CoCl_2{\cdot}7H_2O$	0.15	NH_4^+–N	23.1~57.2
$NaHCO_3$	0.25~0.6	$CuSO_4{\cdot}5H_2O$	0.03	NO_3^-–N	<1
$CaCl_2$	0.01	KI	0.18	TN	25~60
$MgSO_4$	0.05	$MnCl_2{\cdot}4H_2O$	0.06	TP	7.0~8.0
Trace element liquid	0.6 mL/L	$Na_2Mo{\cdot}2H_2O$	0.06	Alkalinity (as $CaCO_3$)	100~600
		$ZnSO_4{\cdot}7H_2O$	0.12		

3.2. The Continuous Flow Test Device

The improved A^2O system test device is shown in Figure 2. The test device was made of plexiglass and is a double-channel plug-flow rectangular reactor with a size of 750 mm × 300 mm × 450 mm with an effective volume of 90 L. It was divided into four reaction sections: the anaerobic section, the pre-anoxic section, the main anoxic section and the aerobic section. The effective volume of each section is listed in Table 3.

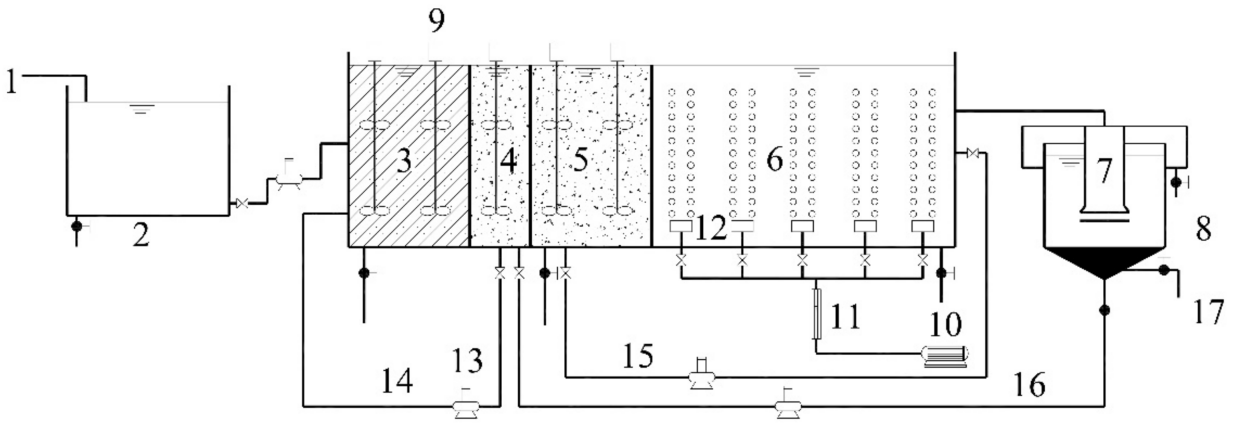

Figure 2. Improved A^2O process flow chart: 1. water inlet; 2. water tank; 3. anaerobic section; 4. pre−anoxic section; 5. main anoxic section; 6. aerobic section; 7. sedimentation tank; 8. water outlet; 9. agitator; 10. air compressors; 11. rotor flowmeter; 12. microporous aerator; 13. constant flow pump; 14. mixed liquid internal circulation; 15. nitration solution internal circulation; 16. sludge reflux; 17. excess sludge.

Table 3. Technical parameters of improved A^2O process.

Reactor	Effective Volume/L
Anaerobic stage	18
The pre-anoxic section	9
The main anoxic section	18
Aerobic stage	45
Sedimentation tank	36

3.3. Test Plan

The experimental plan and operation parameters are shown in Table 4.

Table 4. Operation scheme and related parameters of improved A^2O process.

Test Plan (Number of Days in Operation)	A1 (1~30 d)	A2 (31~60 d)	A3 (61~90 d)	A4 (91~120 d)
ANA series reactor number	2	2	2	2
ANO2 series reactor number	1	2	3	4
SRT(d) [23]	12	12	12	12
Excess sludge discharge/(L/d)	2.29	2.29	2.29	2.29
Inflow (L/h)	10	10	10	10
Sludge reflux ratio	0.5	0.5	0.5	0.5
Ratio of mixed liquid internal circulation	1	1	1	1
Ratio of nitration solution internal circulation	4	4	4	4

Note: ANA represents the anaerobic section, and ANO2 represents the main anoxic section.

The pre-anoxic stage of the improved A^2O process was a single-stage complete mixed reactor, and the aerobic stage was a five-stage complete mixing reactor operating in series. The operating conditions and parameters were unchanged to evaluate the effects of the series of the anaerobic and the main anoxic stages on the efficiency of the nutrient and COD removal and the anoxic phosphate absorption performance. The influent water quality was kept at the same level, with the influent COD concentration of 300 mg/L, TN concentration of 60 mg/L and TP concentration of 8.5 mg/L. The test was conducted for 120 days, and various water quality indicators were detected daily. Statistical methods were used to analyze the measurement results during the stable operation of each test stage, and the number of data points for analysis ranged from 20 to 30.

3.4. Index Detection

During the test, the tested sewage indicators included COD, TP, TN, NH_4^+–N, NO_3^-–N and NO_2^-–N, etc. The water samples were taken out from the anaerobic section, the pre-anoxic section, the main anoxic section and the aerobic section of the reactor, respectively, and then centrifuged at 4500× *g* rpm for 7 min with a centrifuge. The supernatant was taken, and each water quality index was determined using the standard method specified by the state [29].

4. Results and Discussion

4.1. Effect of Different Reactor Stages in Series on Anoxic Phosphorus Absorption

The COD concentration at inlet and outlet water was detected in each experimental stage. During the test, the inlet water quality changed slightly, and the COD was mainly degradable. The COD concentration of the outlet water was lower than 30 mg/L and remained stable, independent of the number of reactors in series in the main anoxic section. Therefore, the COD concentration was not listed.

Figures 3–6 show the change rules of COD, TP, TN, NH_4^+–N, NO_3^-–N and NO_2^-–N concentrations in each reactor in stages, i.e., A1–A4. Taking each reaction section of the improved A^2O process as the system boundary, the material balance analysis was carried out based on the average value of the measured TP and TN concentrations of the mixed liquid, and the TP and TN reaction volumes of each section were calculated (calculation results are shown in Figure 7). It can be seen from the figures that when the number of reactors in series in the main anoxic section was different, the TP concentration of the effluent in the anaerobic section and the amount of phosphorus released in the anaerobic section remained basically unchanged.

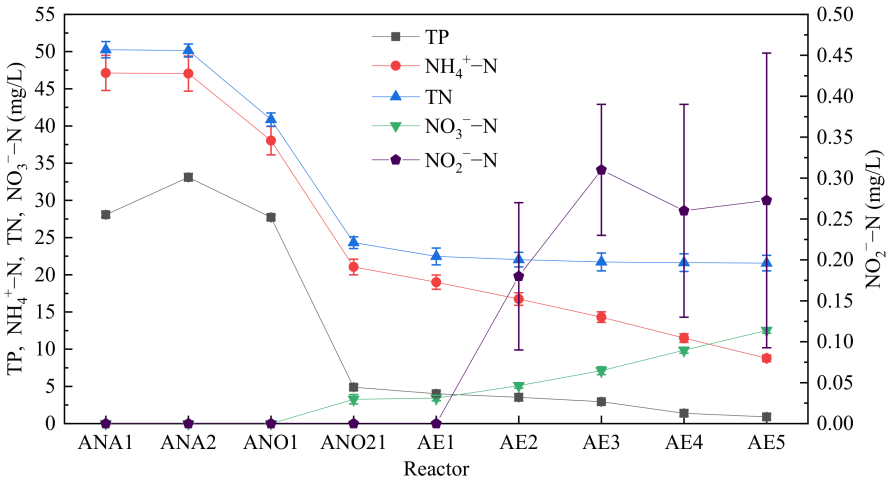

Figure 3. Variation rule of TP, COD, TN, NH_4^+–N, NO_3^-–N, NO_2^-–N concentration in each reactor in A1 stage.

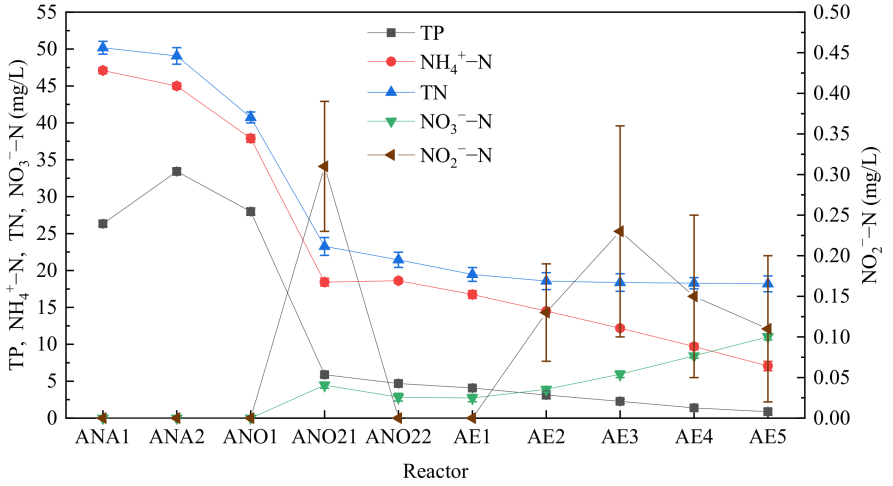

Figure 4. Variation rule of TP, COD, TN, NH_4^+–N, NO_3^-–N, NO_2^-–N concentration in each reactor in the A2 stage.

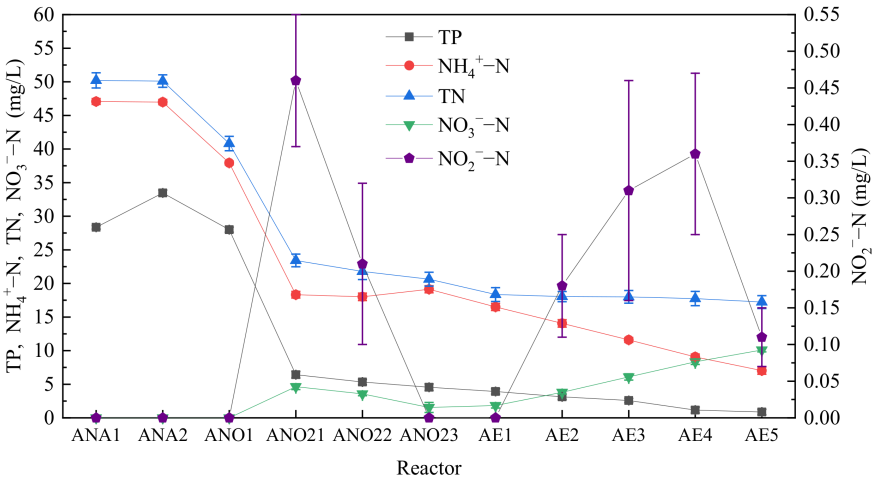

Figure 5. Variation rule of TP, COD, TN, NH_4^+–N, NO_3^-–N, NO_2^-–N concentration in each reactor in the A3 stage.

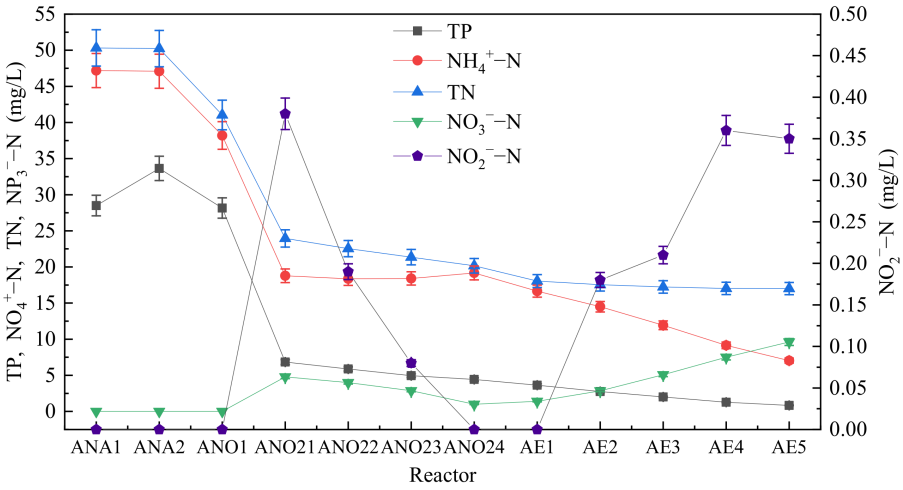

Figure 6. Variation rule of TP, COD, TN, NH_4^+−N, NO_3^-−N, NO_2^-−N concentration in each reactor in the A4 stage.

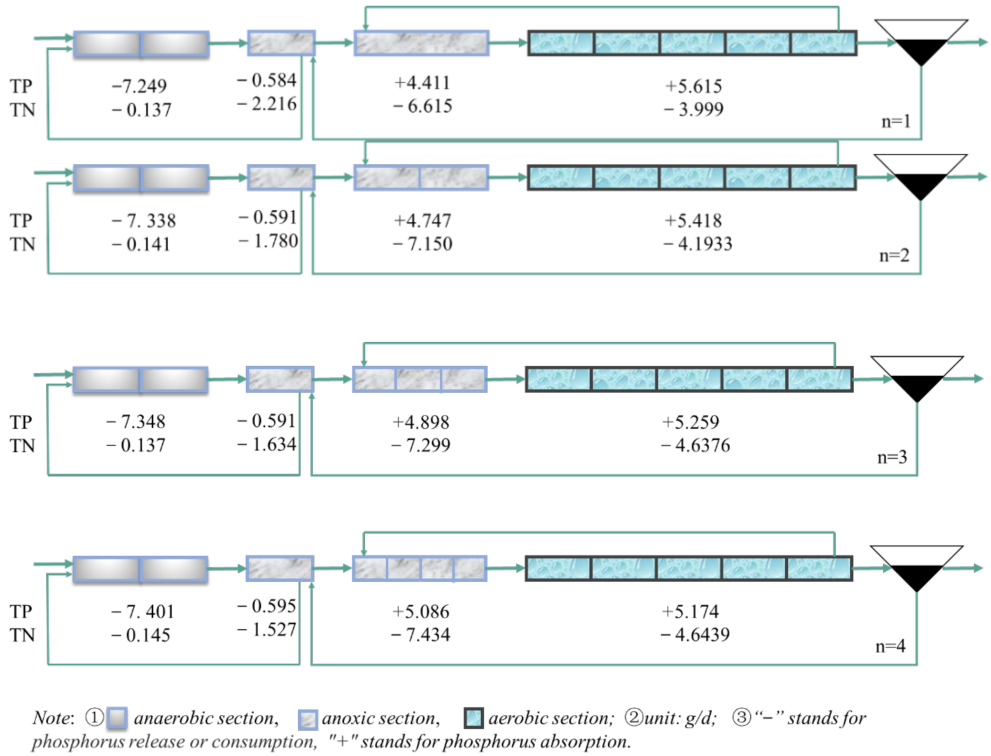

Figure 7. TN and TP reaction amount during the test period.

Note: (1) in the Figures, ANA stands for the anaerobic section, ANO1 stands for the pre-anoxic section, ANO2 stands for the main anoxic section, AE stands for the aerobic section and Arabic numerals at the end stand for the i-stage reactor in series, i = 1, 2, 3, 4; (b) The data are the results of statistical analysis: mean (standard deviation).

In the improved A^2O process, with the increase in the number of reactors in series in the main anoxic section, the concentration of TP in the effluent of the main anoxic section decreased, the amount of anoxic phosphorus absorption increased, and the mass fraction of anoxic phosphorus absorption continued to rise. Each reactor in the series operated in a complete mixed mode. When the reactor was operated at a single stage, the water flow was completely mixed, and the phosphorus absorption power was low. According to the research reports of Lian et al. [14] and Qin et al. [15], if multiple complete mixing reactors were connected in series, the water flow state would be similar to that of horizontal flow

reactors, and the higher the number of reactors in series, the closer to the ideal horizontal flow reactor. Therefore, with the increase in the number of reactor stages in series in the main anoxic section, the higher the anoxic phosphorus absorption power, the faster the anoxic phosphorus absorption rate (see Figure 8 for the anoxic phosphorus absorption rate), and the higher the anoxic phosphorus absorption mass fraction. (When the reactor stages in series of the main anoxic section were one, two, three and four, the anoxic phosphorus absorption mass fraction was 0.44, 0.467, 0.482, and 0.496, respectively.) This helped reduce the demand for carbon sources and improve sewage's nitrogen and phosphorus removal effect with a low carbon-to-nitrogen ratio. In addition, with the increased number of the reactor in series, the reaction kinetics was enhanced, and the reaction rate was faster. The results of this study are similar to the research obtained from treating kitchen wastewater by two reactors in series by Rao et al. [30] using an up-flow anaerobic sludge blanket expanded granular sludge bed (UASB), and also similar to the results obtained by Zhang et al. [31] when dissolving ozonated gases in series with completely mixed reactors.

For the effluent TP concentration of the improved A^2O process, although the effluent TP concentration of the main anoxic section was greatly affected by the number of reactors in series, the system had a good effect on TP removal due to the existence of the subsequent aerobic phosphorus absorption process. The effluent TP concentration was about 0.88 mg/L during the test, and the TP removal rate was maintained at about 90%.

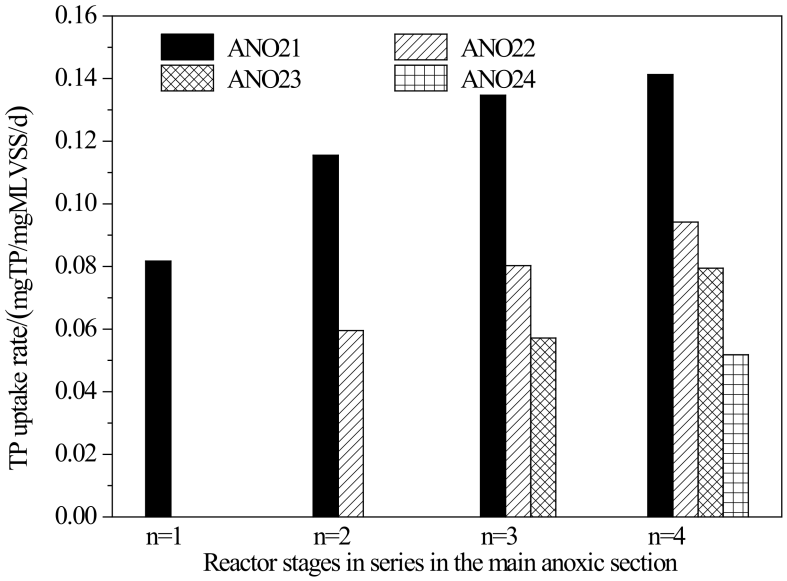

Figure 8. TP absorption rate in the main anoxic reactor at different stages under different reactor stages in series.

Note: ANO2 represents the main anoxic stage, n represents the number of reactors in series, and the last Arabic numeral represents the *i*th reactor in series, i = 1, 2, 3, 4.

4.2. TN Removal Performance under Different Series Reactor Stages

It can be seen from Figures 3–7 that the TN concentration of the system effluent tended to decrease with the increase in the number of reactors in series in the main anoxic section. Furthermore, the total nitrogen in the effluent was mainly NO_3^-–N and ammonia nitrogen. Therefore, the decrease in TN concentration was mainly caused by the decrease in NO_3^-–N concentration. Under the condition that the internal circulation ratio, α, of the nitrification solution remained unchanged when the number of reactors in series was increased from one to two, the denitrification potential of the improved A^2O process was more fully and effectively utilized. The effluent NO_3^-–N concentration was significantly reduced from 12.53 mg/L when n was one, to 9.62 mg/L when n was four, and the removal rate of TN increased from 56.86% when n was one to 65.98% when n was four. However, when

the number of reactors in the series continued to increase to three and four, NO_3^-–N concentration did not decrease significantly. In addition, when the internal circulation ratio, α, of the nitrifying solution remained unchanged, with the increase in the number of reactors in series in the main anoxic section, the reduction power of NO_3^-–N increased, and the denitrification rate accelerated (as shown in Figure 9).

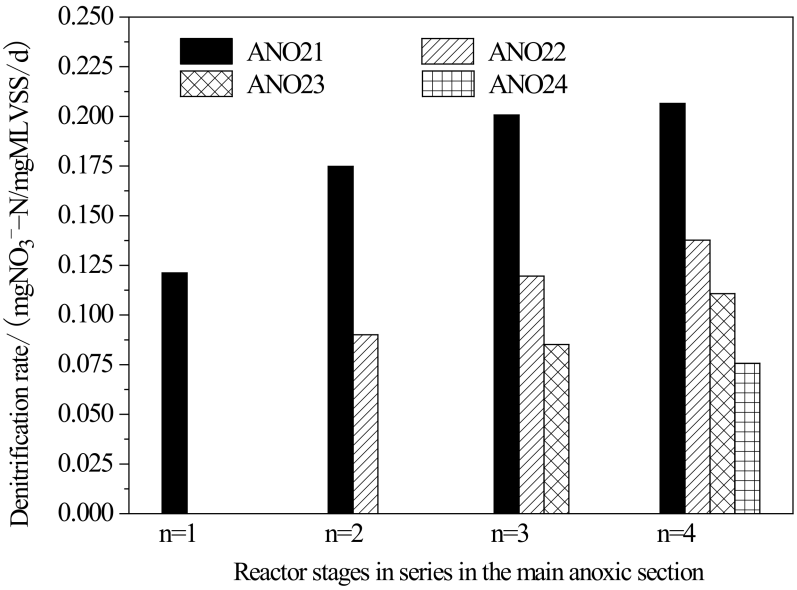

Figure 9. The reduction rate of NO_3^- −N in the main anoxic reactor of each stage under the condition of different series reactor stages.

Note: ANO2 represents the main anoxic stage, n represents the number of reactors in series, and the last Arabic numeral represents the *i*th reactor in series, i = 1, 2, 3, 4.

5. Conclusions

In order to determine the number of reactor stages in series in the main anoxic section of the improved A^2O process, tests were conducted to control the number of reactor stages from one to four, respectively. The results showed that with the increase in the number of reactors in series in the main anoxic section, the TP concentration in the effluent of the main anoxic section was obviously reduced, and the mass fraction of anoxic phosphorus absorption significantly increased. However, due to an aerobic phosphorus absorption process, the system had a good effect on TP removal, and this had little relationship with the series. During the test, the TP concentration in the effluent was about 0.88 mg/L, and the TP removal rate was maintained at about 90%.

Furthermore, under the condition that the nitrifying solution's internal circulation ratio, α, remained unchanged, with the increase in the number of reactors in series in the main anoxic section, the concentration of NO_3^-–N in the effluent of the modified A^2O decreased. As a result, the TN removal effect was improved, the NO_3^-–N reduction power was increased, and the denitrification rate was accelerated. With the increased number of reactors in series, the anoxic phosphorus absorption and denitrification performance was enhanced. In effect, it can reduce the effective volume of the reactor and save construction costs. On the other hand, it can improve the sewage treatment effect, with obvious environmental and economic benefits. Therefore, the main anoxic section of the improved A^2O process can be designed and operated as reactors in series.

The synchronous nitrogen and phosphorus removal system also includes an anaerobic section and aerobic stages. Therefore, in the next step, the contribution of the number of reactors in series to the sewage treatment capacity improvement of anaerobic phosphorus release, aerobic phosphorus uptake, and nitrification reaction can be carried out. Moreover, the influence of reactor stages in series on simultaneous nitrogen and phosphorus removal

can be theoretically improved. Then, the optimal value or recommended range of reactor stages in series in each reaction zone of the simultaneous nitrogen and phosphorus removal process can be proposed.

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