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Enhancing the Stability of Aerobic Granular Sludge Process Treating Municipal Wastewater by Adjusting Organic Loading Rate and Dissolved Oxygen Concentration

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Abstract: Aerobic granular sludge (AGS) application in treating municipal wastewater has been greatly restricted due to its low stability. It has been found that operation parameters have a great impact on stability. The organic loading rate (OLR) and dissolved oxygen (DO) concentration are two very important parameters that impact stability. In this study, the organic loading rate (OLR) and aeration rate were studied to verify their influence on AGS system stability, which is indicated by determining pollutant removal performance, including chemical oxygen demand (COD), ammonia nitrogen, and total nitrogen (TN). The physical and chemical property changes of AGS and the effects of pollutant removal during the formation of AGS were systematically investigated. The AGS was formed after about 25 days and remained stable for about 45–50 days. The AGS was light-yellow globular sludge with an average particle size of 1.25 mm and a sludge volume index (SVI) of 33.9 mL/g. The optimal condition was obtained at an OLR of 4.2 kg COD/m^3·d, aeration rate of 4 L/min, and a hydraulic retention time (HRT) of 4 h. The corresponding removal efficiencies of COD, ammonia nitrogen, and TN were 94.1%, 98.4% and 74.1%, respectively. The study shows that the AGS system has great potential for pollutant removal from wastewater.

Keywords: aerobic granular sludge; municipal wastewater; AGS system; organic loading; dissolved oxygen

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1. Introduction

Granular sludge (GS) can be considered a special type of biofilm [1]. It is a denser, larger and better-bodied aggregation of microorganisms in biological wastewater treatment systems. It is generally generated under appropriate environmental conditions. There are two types of GS: aerobic granular sludge and anaerobic granular sludge, which are classified based on the electron acceptor and the metabolism process. The aerobic granular sludge (AGS) process has received great attention due to its ease of operation [2,3].

The AGS process has many advantages over traditional activated sludge processes in wastewater treatment, including its regular appearance, compact conformation, superior sedimentation speed, forceful impact load resistance, and high flexibility [4,5]. In addition, the AGS process achieves good technical and economic performance [6,7]. The AGS process is not only effective for high concentrations of chemical oxygen demand (COD) removal but also could absorb heavy metals when it is used to treat industrial wastewater [8,9]. In addition, it has been found that AGS has the ability to simultaneously remove nitrogen and phosphorus, which has broad potential in domestic sewage treatment [10,11].

Therefore, research on AGS process application in domestic sewage treatment has gradually gained increasing interest in recent years. However, so far, the AGS process has only been applied to treat industrial wastewater [12–14], and the use of the AGS process for treating actual domestic wastewater is limited, which limits the further promotion and application of AGS technology.

The major problem with AGS used to treat municipal wastewater is poor stability. Several parameters have been found to impact the stability of the AGS process when it is used to treat municipal wastewater. The formation of AGS has a great impact on process stability. Several studies have found that the formation of AGS is affected by many factors, including organic loading, hydraulic shearing stress, settling time, and feast-famine period [15–18]. High-strength gas, hydraulic shear force caused by aeration or mud-water collision, large aspect ratio, and ideal flow process provide better conditions for the formation of AGS [19,20]. Furthermore, studies have shown that a shorter settling time can stimulate the secretion of microbial extracellular polymeric substances (EPS), which is important for improving the characteristics of AGS and promoting the formation of AGS [21]. The slow settling speed can be achieved through the sequencing batch reactor (SBR) operation mode [22–26]. The bacterial growth rate has also been reported to impact the formation of AGS, and the growth rate can be increased under high organic loading [27,28]. In fact, all the parameters that impact AGS formation can be induced by varying the organic loading rate and the aeration rate, as they can directly or indirectly influence the feast-famine phase and shearing force.

Therefore, in this study, the AGS process was employed to treat municipal wastewater. The impact of the organic loading rate and the aeration rate on the stability of AGS has been investigated. Optimal conditions have been obtained. The relationship between AGS stability and treatment performance has been explored.

2. Materials and Methods

2.1. Reactor Set-Up and Operation

The reactor used had a total effective volume of about 4.4 L with a height of 1150 mm and an inner diameter of 80 mm. The seven sampling ports with a diameter of 10 mm were evenly located on the side of the cylinder, with a distance of 125 mm. Moreover, a 10 mm diameter water inlet was added on the opposite side in parallel with the bottom sampling port. An aeration disk (a diameter of 80 mm) fixed on the bottom was connected with a cylinder by a flange. The SBR process contains five basic processes: feeding (15 min), aeration (200 min), settling (15 min), drainage (5 min) and standby (5 min), which was controlled by a time-control system. The wastewater was filled through a peristaltic pump from the bottom of the water inlet.

2.2. Wastewater and Cultivate Sludge

During the AGS formation period, the feeding wastewater was a mixture of synthetic wastewater with 5~10% real domestic sewage wastewater. The composition of the feeding wastewater is provided in Table 1.

Table 1. The characterization of the treated wastewater.

Components	Concentration (mg/L)	Components	Concentration (mg/L)
COD	350~560	BO_3^{3-}	0.05
NH_4^+-N	59.2~90	Cu^{2+}	0.05
TP	3.9~5.5	Zn^{2+}	0.05
TN	0~2.5	Al^{3+}	0.09
NO_3^-	60.4~91.7	Co^{2+}	0.05
SS	20~65	Mn^{2+}	0.05
Ca^{2+}	30	$Mo_7O_{24}^{2-}$	0.05
Mg^{2+}	25	Ni^{2+}	0.09
pH	6.3	Fe^{3+}	0.05

The seed sludge was obtained from an aerobic section of a wastewater treatment plant in Shenzhen. The sludge volume index (SVI) of the inoculated sludge was about 95 mL/g and was incubated with synthetic water for one week before utilization.

2.3. AGS Process Start-Up

It is known that a high C/N ratio could shorten the AGS formation time [29]. After one week of cultivation, the sludge color gradually became dark brown with an SVI of about 100 mL/g, and then it was transferred to the SBR reactor with an initial sludge concentration of 5256 mg/L to form AGS. The AGS process then started with a hydraulic retention time (HRT) of 4 h.

2.4. Pollutant Removal with AGS from Wastewater

The operation was divided into three stages: stage I (0 d–10 d), stage II (10 d–45 d), and stage III (45 d–57 d). In the three stages, the HRT is the same, which is 4 h, but the time division, including feeding time (15 min), aeration time (200 min for stage I, 205 min for stage II, 210 min for stage III), settling time (15 min for stage I, 10 min for stage II, 5 min for stage III), drainage time (5 min), and standby time (5 min), is different. The wastewater was fed to the reactor from the inlet and then aeration was provided. The microorganism in the AGS starts to degrade COD and ammonia under aerobic conditions. Part of the COD was turned into CO_2 and the rest was turned into a new microorganism, which led to COD removal. Ammonia nitrogen was converted to nitrate by nitrification bacteria. Since the settling period, the system gradually became anaerobic; thus, some of the nitrate was transferred to nitrogen gas and total nitrogen was removed. However, due to the low COD concentration in the anaerobic period, TN removal was limited.

2.5. The Impact of OLR and Aeration on AGS Stability

After the AGS was formed and the system was stable, the OLR was adjusted by the reduction or addition of glucose to the wastewater. The final COD was set at 300 (corresponding to 1.8 kg COD/m^3·d), 500 (corresponding to 3 kg COD/m^3·d), 700 (corresponding to 4.2 kg COD/m^3·d), and 900 mg/L (corresponding to 5.4 kg COD/m^3·d) to study its impact on the treatment performance stability. After optimal OLR was obtained, the aeration rate (2 L/min, 4 L/min, 8 L/min) was optimized as well. Samples were taken during the operation, and pollutant removal efficiency was evaluated.

2.6. Analytical Methods

The concentrations of COD, ammonia (NH_4^+-N), nitrate (NO_3^--N), nitrite (NO_2^--N), total nitrogen (TN), total phosphorus (TP), mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), sludge volume (SV) and sludge volume index (SVI) were measured according to Standard Methods for the Examination of Water and Wastewater (APHA, 2005). Dissolved oxygen (DO) and pH were determined using a DO meter (Visay YSI550A, USA) and a pH meter (PHS-2F, Shanghai, China), respectively.

The particle size distribution of the AGS was measured by a laser particle size analyzer (Malvern Malvern, Mastersizer 2000 series, England). The AGS sedimentation rate was determined using the static sedimentation method [30]. EPS was extracted using the hot extraction method [31,32]. A modified rapid Lowry method protein content determination kit (PRL002000, Shanghai, China) was used to determine the protein concentration of the AGS. Glucose was used as a standard, and the phenol-sulfuric acid method was used to determine the polysaccharide concentration. A scanning electron microscope (SEM) (Hitachi S4700, Japan) was used to observe the surface morphology of the granular sludge. DNA extraction was performed by using FastDNA®SPIN kit (MP Biomedicals, USA) for the soil kit.

3. Results and Discussion

3.1. Formation of AGS during the Start-Up Stage

Particle size distribution was measured at 0 d, 20 d, 40 d, and 60 d. It was found that the particle size was 40 to 100 μm with average size of around 60 μm initially at 0 d. Gradually, granular sludge was formed. At 20 d, a small amount of granular sludge was observed and the average particle size was around 160 μm. The granular sludge was in greater portion than sludge floc and the average size was about 800 μm at the 40th d. Mature AGS was obtained at 60 d and the average size was 1200 μm. This suggests that the AGS formed around 40 d.

According to the variation of MLSS, MLVSS and SVI (Figure 1), it can be seen that the initial sludge concentration in the reactor was 5256 mg/L, and SVI was 94.8 mL/g. Due to the sudden decrease of settling time to 15 min from 30 min, a large amount of sludge with poor settling ability was discharged from the reactor and thus resulted in a sudden decrease in sludge concentration on the following days (10th–42th).

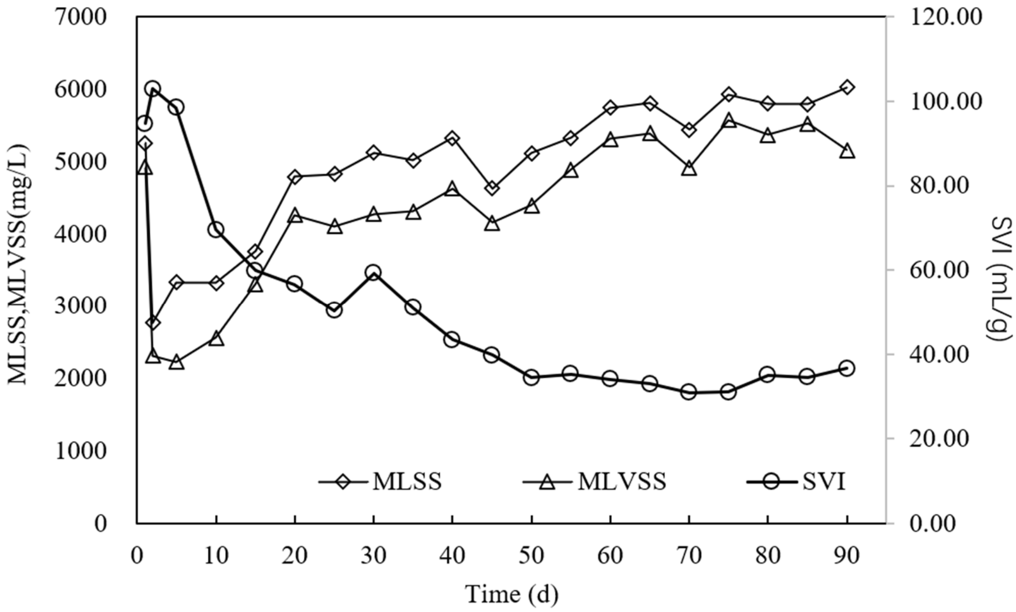


Figure 1. Variation of MLSS, MLVSS and SVI in AGS formation and operation.

With the selection of AGS, the sludge concentration in the reactor gradually increased and SVI continuously decreased. On the 25th day, the increasing rate of MLSS began to slow down, which represented a reduction in the microorganism growth rate. In addition, light yellow spherical particles were found in the reactor, which revealed the formation of AGS. Moreover, SVI remained stable while MLSS slowly increased after the 50th day, with MLSS and MLVSS values of 5664 mg/L and 5172 mg/L, respectively. This indicates the successful cultivation of AGS. The MLVSS/MLSS has been maintained above 0.85, which suggests that the system was in good bioactivity. The SVI decreased to 33.9 mL/g from the initial 94.8 mL/g. This indicates that the AGS has excellent settling performance.

3.2. Pollutant Removal by AGS Process

Pollutant removal efficiency was an important indicator of microbial activity and process capacity. The removal efficiency of COD in the start-up period is shown in Figure 2a. At the initial start-up stage, microbial adaptability was poor and the unstable degradation rate of COD was observed. The removal efficiency of COD in stage I was below 80%. In stage II, the sludge concentration increased to about 4600 mg/L with the growth of microorganisms. The COD concentration in the effluent on the 28th day met the demand of the first A grade of the China National Wastewater Discharges Standard [33].

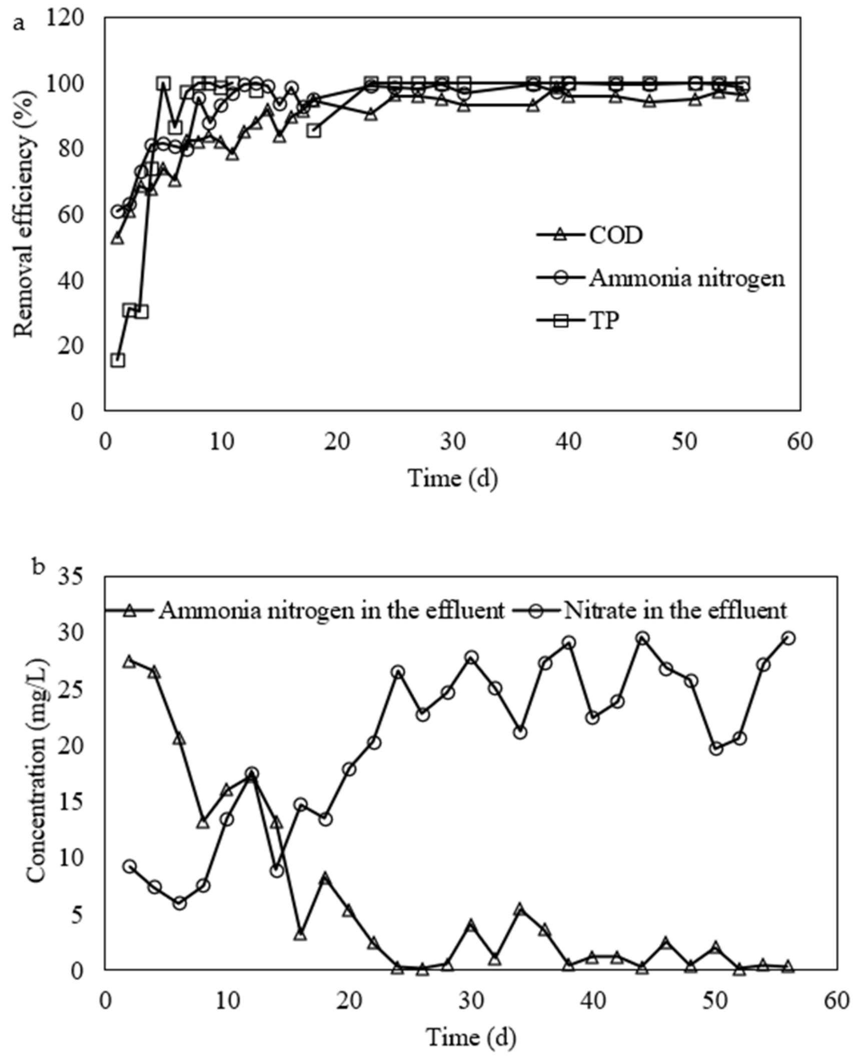


Figure 2. Pollutant concentration variation and removal efficiency. (**a**) the pollutant removal efficiency; (**b**) concentration variation of ammonia and nitrate in the effluent.

The removal of NH_4^+-N in start-up process is shown in Figure 2a. Initially, the sludge settling ability was poor; high settling time (15 min) is not suitable for AGS selection and thus caused poor ammonia nitrogen removal. It fluctuated between 60% and 80%. With the reduction of sedimentation time, the removal rate of NH_4^+-N has increased to above 90% due to the granularization of AGS. The removal rate of NH_4^+-N remained above 97% when the settling period was 5 min.

The production of NO_3^--N is shown in Figure 2b. It can be seen that even though this system has excellent removal in NH_4^+-N, accumulation of nitrate occurred during the entire start-up process. At the beginning, the average concentration of NO_3^--N in the effluent was 10.0 mg/L and reached 26.6 mg/L at 26th day. When it stabilized, the nitrate in the effluent remained above 20 mg/L. This demonstrated the poor denitrification effect and the low activity of denitrifying bacteria. This could be because the aeration in the reactor was high, the size of the AGS was small, and the efficiency of dissolved oxygen transfer was high. As a result, the anaerobic area is so small that denitrifying bacteria cannot obtain a suitable microenvironment for growth.

Phosphorus removal efficiency was unstable and ranged from 16.0% to 70.8% at the early stage of startup due to the low activity of polyphosphate bacteria (Figure 2a). On the 30th day, the effluent total phosphorus (TP) concentration was below 0.5 mg/L with a removal rate of 90.7% and reached 97.9% later. Phosphorus concentration is greatly associated with SS. In addition, the high removal efficiency of phosphorus could also explain the low denitrifying bacteria activity in this system due to competing with the polyphosphate bacteria for nutrition sources.

3.3. Effect of Organic Loading Rate in an AGS System

The organic loading rate (OLR) is one of the most important parameters for the stable operation of AGS. Studies have shown that when the OLR is very low, the growth rate of microorganisms is slow and aggregates are difficult to form. However, when the OLR is high, reaching 6–9 kg/m^3·d, microorganisms tend to overgrow. The AGS forms a loose layer, which results in poor settling performance. Under suitable OLR conditions, it is easy to form an AGS with a compact structure and reaches a high biomass concentration in the system. In general, the bacterial growth rate can increase with an increase in OLR.

In this study, after AGS was formed and stable, the OLR was first set at 1.8 kg COD/m^3·d for 14 d, and then 3 kg COD/m^3·d for the next 14 d, followed by 4.2 kg COD/m^3·d for another 14 d, and finally 5.4 kg COD/m^3·d for the next 12 d. The total operation time to investigate the OLR impact was 56 d. COD removal under different OLR conditions is shown in Figure 3a. The removal rate of COD remained above 88.0% with varying OLR between 1.8 and 4.2 kg COD/m^3·d. When OLR was at 5.4 kg COD/m^3·d, COD concentration in the effluent increased to 130 mg/L at the first day. This might be because the system is impacted by a sudden increase in organic loading. Thereafter, it gradually decreased. The average removal rate reached 91.8% on the 3rd day, with the effluent concentration around 50 mg/L. This is due to the adaption of the AGS to the influent concentration.

The impact of OLR on the removal of NH_4^+-N is shown in Figure 3a. The initial NH_4^+-N in the influent was about 70 mg/L. Therefore, the variation of OLR (COD) means the variation of C/N ratio. The corresponding C/N ratio was around 4.28, 7.14, 10 and 12.86 with COD concentrations of 300 mg/L, 500 mg/L, 700 mg/L and 900 mg/L. It was observed that a low C/N ratio enhanced the removal of NH_4^+-N. With the increase of C/N ratio, the removal efficiency of NH_4^+-N decreased. At a high C/N ratio, the carbon source is sufficient, which leads to competition between heterotrophic microorganisms and autotrophic nitrifying bacteria for dissolved oxygen [34]. The inhibited growth rate of nitrifying bacteria resulted in a weak nitrification reaction. The removal efficiency of NH_4^+-N decreased with C/N ratio less than 7.14.

It was also exhibited that the concentration of NH_4^+-N in the effluent was higher than 5 mg/L, and the average removal rate was 89.5%, with a C/N ratio of 4.28 (Figure 3a). With the increase of C/N ratio, the nitrification reaction gradually improved and the removal efficiency of NH_4^+-N reached 94%; however, the concentration of NH_4^+-N in the effluent would increase when C/N ratio was greater than 10. Because it is more conducive to a sufficient carbon source to enhance the growth of heterotrophic bacteria, and thus under competition conditions, nitrifying bacteria were inhibited. Overall, the average concentration of NH_4^+-N in the effluent was higher than 5 mg/L with the average removal rate of 92.6%.

The removal rate of TN increased with an increase in OLR (Figure 3a). The removal efficiency of TN was only 56.2% at C/N ratio of 4.28. This could be because the carbon source in this system was insufficient to satisfy the needs of granular sludge, which is necessary for bacterial growth. Inside each AGS reactor, organic matter is quickly consumed in the aerobic layer, cannot reach the anoxic layer, and thus inhibits growth. The metabolism of denitrifying bacteria belongs to facultative anaerobes and then results in poor TN degradation. With an increase in OLR, the removal efficiency of TN gradually increased. Since the OLR was 4.2 kg/m^3·d, the removal rate of TN went up to 75.6%. The average concentration of TN in the effluent was 17.67 mg/L. As the OLR increased to 5.40 kg/m^3·d and the C/N ratio rose to 12.86, the TN removal rate decreased. This may be caused by the reduction of nitrifying bacteria, which leads to the increase of TN concentration.

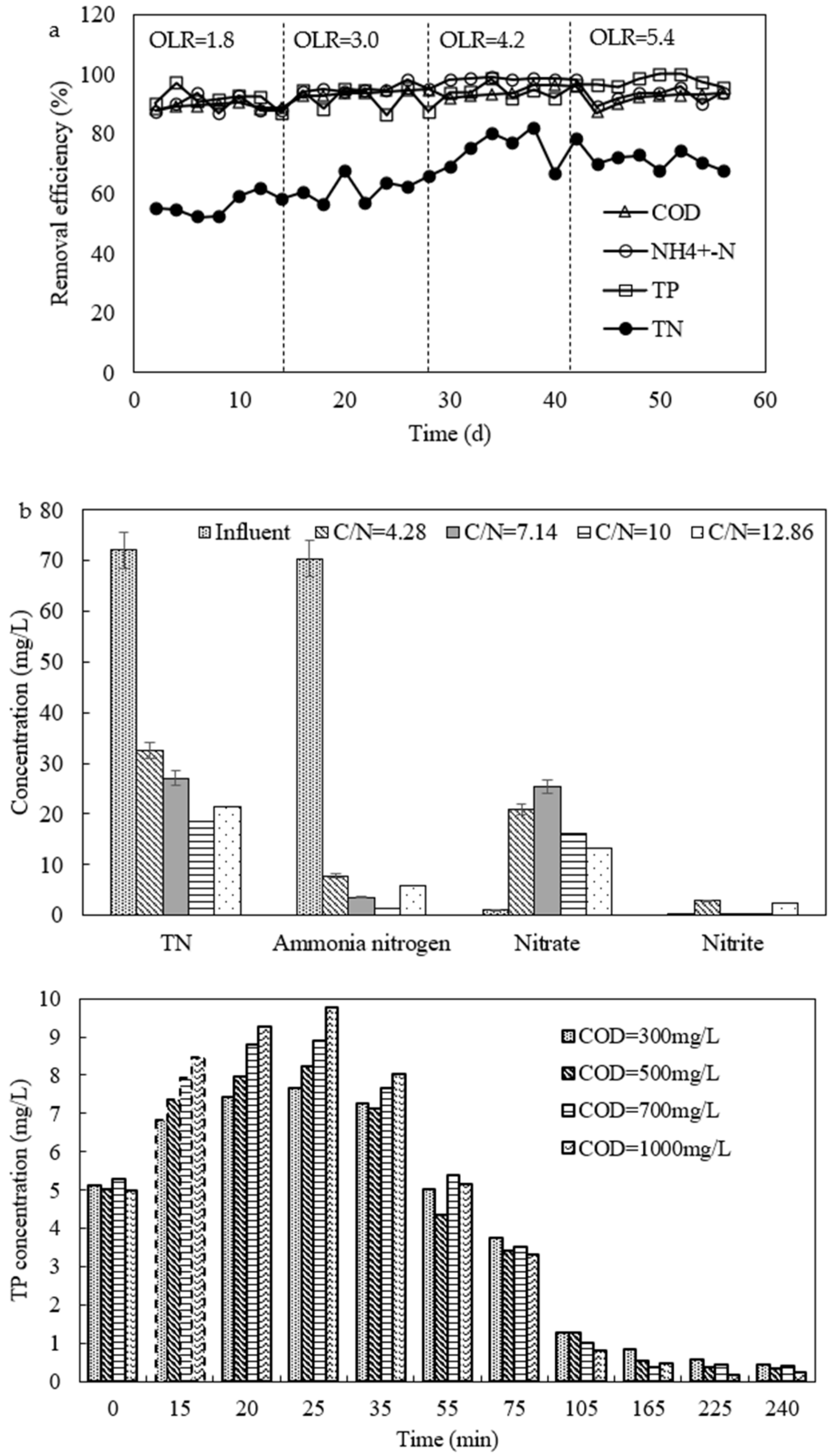


Figure 3. Effects of different organic loadings on COD removal, ammonia nitrogen removal, total nitrogen removal and phosphorus removal (the OLR unit is kg COD/m^3·d). (**a**) pollutant removal efficiency; (**b**) pollutant concentration in the effluent; (**c**) TP concentration variation in the effluent with the variation of influent COD.

Figure 3b shows that, under different conditions, NO_3^--N is one of the major components of TN in effluent. With the increase of C/N ratio, the nitrification reaction gradually enhanced, and the concentration of NO_3^--N in the effluent increased. Additional carbon sources in the system can be used for denitrification. Therefore, while the concentration of TN gradually reduced, the effluent NO_3^--N gradually reduced as well. But when C/N ratio continued to increase to 12.86, the concentration of TN increased and NO_3^--N decreased.

The phosphorus in the influent was almost constant at 5 mg/L, but the concentration of COD in the influent changed, resulting in an increase in COD/TP. Different OLR had little effect on the removal of phosphorus, and effluent phosphorus content was basically below 0.5 mg/L. The variation of phosphorus concentration under different OLR is shown in Figure 3c. At the initial stage of aeration, the phosphorus concentration increased under different conditions. Since microorganisms in the system were starving at this stage, COD was consumed in the meantime. Most of the COD was degraded by heterotrophic bacteria in the outer layer of the granular sludge, and a large amount of DO was consumed simultaneously. The DO could not reach the inner layer where the polyphosphate bacteria were located. In the anaerobic stage and early aeration stage, nutrients are sufficient. At this time, the polyphosphate bacteria released intracellular polyphosphate into the system through hydrolysis and used the absorbed organic matter to synthesize PHB. As the reaction went on, the carbon source in the system decreased rapidly, and the DO reached the area where the PAOs were presenting. On the other hand, the progress of nitrification increased the concentration of nitrate and nitrite in the system. Under this condition, sufficient electron acceptors and carbon sources or energy storage substances in cells could be oxidized and decomposed to enable phosphorus absorption.

Overall, under the conditions 4.2 kg/m^3·d of OLR and C/N ratio of 10, AGS could maintain better stability, higher activity, and better removal effect of various pollutants compared with other situations.

3.4. Effect of Dissolved Oxygen (DO) Concentration

The degradation rate of organic matter and the removal of nitrogen and phosphorus by microorganisms in this system were affected not only by organic loading but also by DO. The effect of aeration on pollutant removal performance and sludge stability has been examined under optimal organic loading with different aeration rates (2 L/min, 4 L/min, 8 L/min corresponding to the superficial velocity of 0.66, 1.33 and 2.65 cm/s). Figure 4 shows the variation in DO concentration in the SBR column under different aeration intensities.

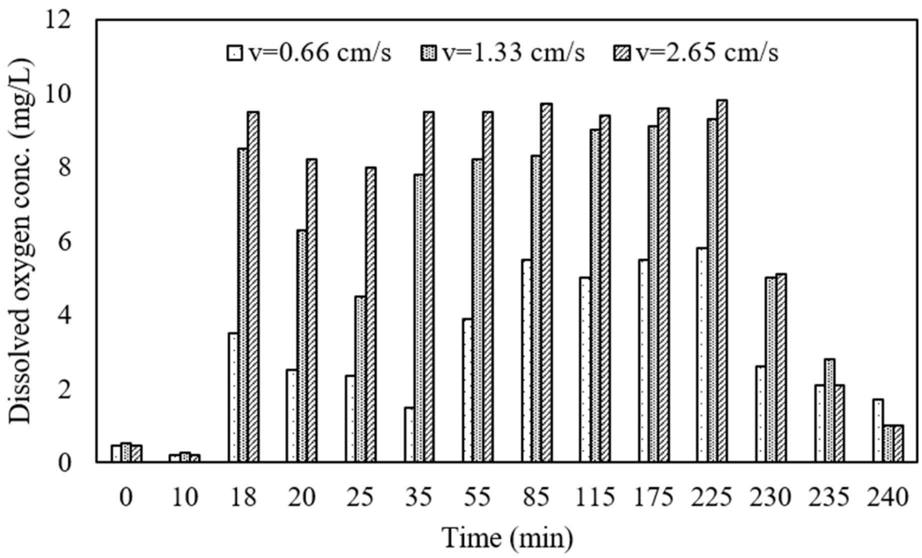
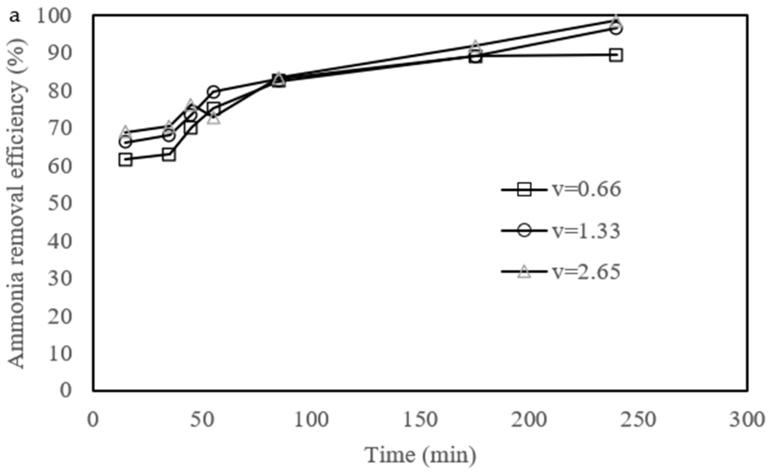
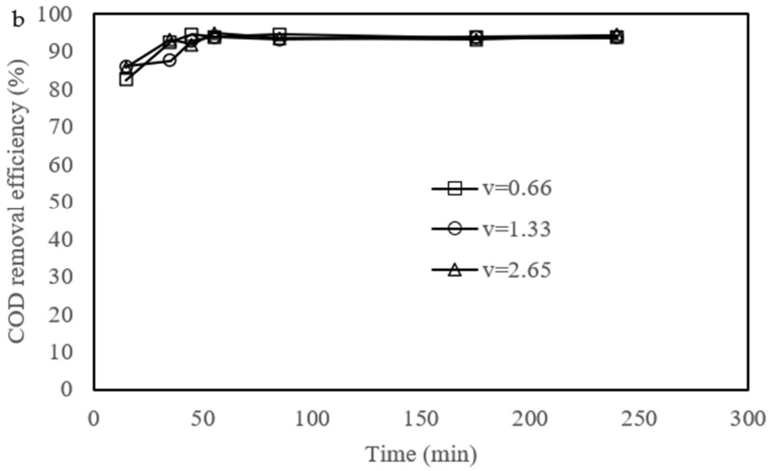


Figure 4. Dissolved oxygen concentration in the reactor at different aeration levels.

The concentration of DO was maintained at around 3.7 mg/L under an aeration volume of 2 L/min at the later period of the aeration process (Figure 4). The concentration of DO was maintained at about 8.6 mg/L under an aeration rate of 8 L/min. During the operation, the concentration of DO was increased within a short period at the beginning of aeration, then declined for a short while, and finally was stable after gradually increasing. Compared with other aeration conditions, it may be relevant to the stages of starvation and hypoxia of microorganisms. In the early stage of aeration, microorganisms quickly absorbed oxygen to catabolize organic matter, which caused a rapid decrease in the DO concentration.

The concentration of COD in the AGS process under different aeration conditions is shown in Figure 5a. COD rapidly decreased at the first 15 min, regardless of the aeration level. The COD concentration in the effluent was basically stable after 35 min.





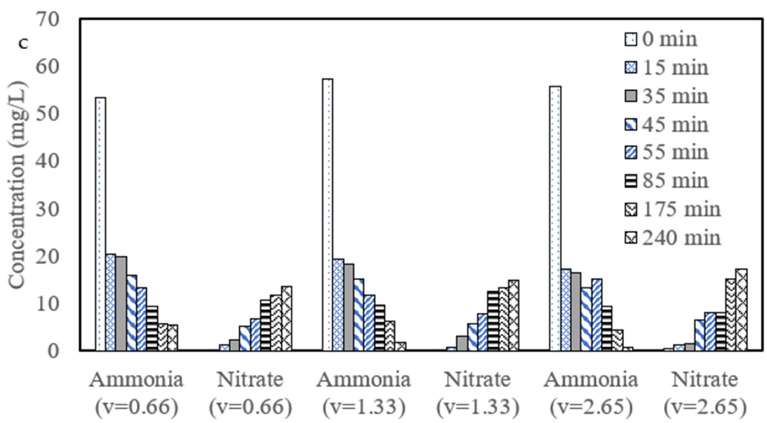


Figure 5. *Cont.*

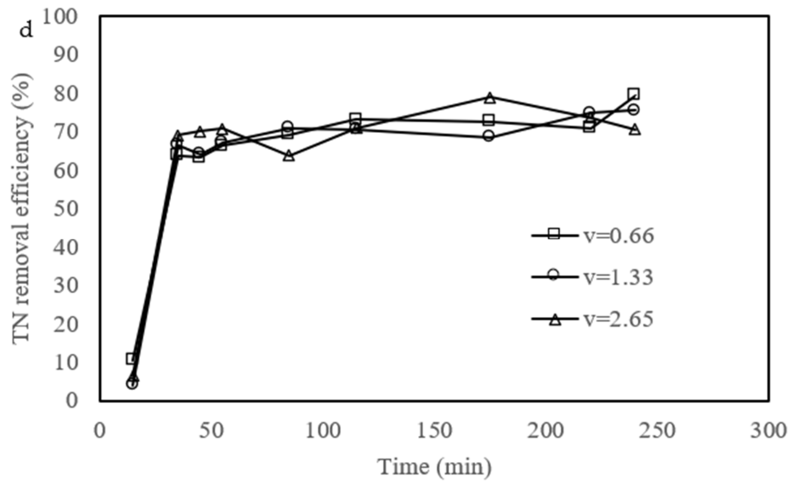


Figure 5. Effect of aeration on (**a**) COD removal; (**b**) ammonia nitrogen removal; (**c**) total removal; (**d**) ammonia nitrogen and nitrate concentration variation.

With the concentration increase of COD in the influent, COD degradation slowed. The sudden increase in COD concentration in the influent could increase osmotic pressure. When it exceeded a certain limit, it affected microorganisms and thus caused low COD degradation efficiency. It was observed that an increase in aeration can enhance the COD removal effect to a certain extent, but the effect was very slight. Therefore, it can be concluded that an aeration rate of 2 L/min could meet the required DO concentration for the normal growth of microorganisms.

Figure 5b shows the effects of aeration on the removal of NH_4^+-N and NO_3^--N in a cycle. At the initial stage of aeration, the degradation rate of NH_4^+-N was high. The removal of NH_4^+-N were 61.83%, 66.21%, and 68.96% at aeration rates of 2 L/min, 4 L/min, 8 L/min, respectively, within the first 15 min. However, the concentration of NO_3^--N only increased by 4.22, 5.75, and 4.34 mg/L which was far less than NH_4^+-N removal. In addition, no nitrite concentration was detected in the system. This indicates that the removal of NH_4^+-N mainly was the biological adsorption within the first 15 min but not nitrification.

After the adsorption process, AOB in sludge continued to oxidize NH_4^+-N in sludge and NO_3^--N in the system gradually increased (Figure 5c), which resulted in the decrease of NH_4^+-N. At the end of the reaction, the degradation rates of NH_4^+-N were 89.8%, 96.8%, and 98.6%, respectively, and the NH_4^+-N concentration in the effluent met the first A grade of the national wastewater discharge standard.

Furthermore, the accumulation of NO_3^--N increased with an increase in the aeration rate (Figure 5c). It has also been reported by others as well [25]. When the aeration rate reached 8 L/min, accumulation of NO_3^--N occurred, and its concentration in the effluent reached 17.25 mg/L. When DO in the system was high, DO could reach a deeper layer of granular particles, which led to the reduction of the anoxic environment inside the AGS.

The degradation effect on TN under different aeration conditions is shown in Figure 5d. TN was rapidly reduced in the system (about 35 min). This could be due to the AGS biosorption of NH_4^+-N in the initial period, resulting in a rapid decrease in TN concentration.

After the biosorption process, the removal rate of TN first showed a rapid decrease and then a slow decrease. The concentration of organic matter in the system was sufficient at an early stage, but the concentration of NO_3^--N was small; however, the denitrifying bacteria growing speed was relatively fast. With the progress of the reaction, the organic matter in the system decreased rapidly and soon fell under 50 mg/L. At this time, denitrification consumed an internal carbon source. As the reaction proceeded, the removal rate of TN decreased slightly. The aeration rate did not impact TN removal. This suggests that 2 L/min aeration is sufficient to maintain TN removal.

The aeration rate has a great impact on AGS process performance. The 2 L/min aeration rate could achieve stable pollutant removal. This suggests that the aeration rate is significantly important in maintaining the stability of AGS.

4. Conclusions

The cultivation of AGS has been successfully achieved in the laboratory. Sludge granulation was achieved in the SBR reactor within 25 d. The AGS was light yellow spherical with an average particle diameter, SVI and extracellular protein of 1.25 mm, 33.9 mL/g and 185.86 mg/g MLSS, respectively. Two important parameters, including OLR and aeration rate, have been found to have a significant impact on AGS stability to remove pollutants. High pollutant (COD, NH_4^+-N, and TDP) removal efficiency has been achieved under optimal OLR and aeration rates.

The study revealed that the AGS process could be used for municipal wastewater treatment but maintaining sludge in granular during the process by providing proper OLR and aeration rate is very important for guaranteeing the process performance. However, there is a problem with TP removal from wastewater due to the SS escaping from the reactor; therefore, before the application of the AGS process in practice, it needs to solve the SS escaping problem.

Author Contributions: J.P. prepared some parts of the manuscript draft and performed some experiment. L.Z. and Q.W. performed the experiments and analyzed samples. W.S. and Z.W. provided suggestions while encountering problems. J.L. supervised the experiment direction. X.Z. designed the experiment, performed the literature review on the AGS process for municipal wastewater treatment, interpreted the obtained results, and revised the manuscript. F.Y. performed the sample analysis. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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