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Editorial

Environmental Protection through Aerobic Granular Sludge Process

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Aerobic granular sludge (AGS) represents a significant advancement in wastewater treatment technology. This innovative approach differs from traditional methods by utilising self-immobilised biofilms without the need for carrier media. The distinct features of AGS, such as its compact three-dimensional structure and larger size, endow it with remarkable sludge settling capabilities and enhanced biomass retention within reactors. One of the key benefits of AGS is its resilience against toxic compounds and its ability to adapt to fluctuating organic loading rates. This robustness is largely due to the unique physical and biological properties of the granular sludge. Additionally, the use of aerobic granular sludge in treatment plants offers considerable operational advantages by a significant reduction in spatial footprint—up to 75%—and lower energy consumption by 30–60% compared to conventional activated sludge (CAS) systems. The combination of these advantages posits aerobic granular sludge as a highly promising technology as either an alternative to CAS for wastewater treatment or to intensify CAS for better performance.

Aerobic granular sludge technology was first reported in 1991 for municipal sewage treatment in an aerobic upflow sludge blanket reactor [1], similar to an upflow anaerobic sludge blanket reactor for anaerobic granules, but this paper did not gain much attention due to the instability of the formed granules. The granulation of sludge in sequential batch reactors (SBRs) reported in 1997 [2] ignited intensive research and rapid commercialisation of AGS branded as Nereda® by Royal HaskningDHV (Amersfoort, The Netherlands). Despite this, adapting existing continuous-flow CAS systems for SBR-based AGS remains challenging. To address this, sludge intensification processes were developed in continuous-flow reactors by adding additional hydrocyclone separation to enhance sludge settleability through partial sludge granulation. Currently, approximately 60 Nereda® wastewater treatment plants and a few intensified plants exist; however, the full-scale adoption of AGS technology has not reached the anticipated levels after 25 years of intensive research and development. This underscores the need for further advancements and wider application in diverse scenarios, which is the focus of the Special Issue titled "Environmental Protection through Aerobic Granular Sludge Process".

The contents of this Special Issue cover a variety of studies on a range of topics of aerobic granular sludge, which can be classified into categories based on the types of wastewater for treatment, nutrient removal, long-term stability of granule-based reactors, and full-scale granular sludge application. In addition, the Special Issue includes a review paper on the waste granular sludge treatment that full-scale WWTPs have to deal with.

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Treatment of Various Types of Wastewater with Aerobic Granules

Aerobic granular sludge offers advantages over suspended sludge in retaining slow-growing bacteria such as nitrifying bacteria and polyphosphate accumulating bacteria (PAO) for nutrient removal due to its high biomass retention and high actual sludge retention time (SRT). This is crucial for efficient nutrient removal. Zhang et al. reported on the treat of low-strength wastewater with 400 mg/L COD and 100 mg/L NH_4^+-N, respectively, i.e., a COD/N (C/N) ratio of 4:1, for granulation and nutrient removal. It was found that NH_4^+-N removal efficiency reached almost 100%, indicating excellent nitrifying performance. Liu et al. [3] introduced a novel and effective method to rapidly form nitrifying granules, which was to form heterotrophic granules with a fast granulation strategy [4] followed by converting heterotrophic granules into nitrifying granules by reducing COD concentration to 0 while simultaneously increasing NH_4^+-N concentration from 50 to 850 mg/L. Consequently, granules capable of treating ammonia-rich wastewater with 850 mg/L NH_4^+-N were formed within 37 to 49 days at temperatures of 21 °C and 32 °C, respectively. Morgan et al. explored the treatment of nutrient rich side-stream using synthetic centrate or diluted centrate with low COD and a low COD/N ratio for nitrification and denitrification. By adding acetate as the external carbon source, aerobic granules achieved over 90% COD and over 80% P removal without NOx-N accumulation when COD/N ratio was higher than 11 mg sCOD/mg NH_4^+-N. An et al. conducted nitrification in GMBR fed with wastewater containing 50 mg/L NH_4^+-N and without COD, and it was found that excellent nitrification was achieved. Iorhemen et al. focused on phosphorus removal via enhanced biological phosphorus removal (EBPR) using granular sludge for wastewater with an influent COD of 2000 (day 1–27) and 1000 mg/L (day 28–84) and a COD/N/P ratio of 100:5:1. Three different feeding strategies, anaerobic slow feeding (R1), pulse feeding followed by anaerobic mixing (R2), and pulse feeding (R3), were studied. The results showed that R1 and R2 achieved significantly higher phosphorus removal (97.6% for R1 and 98.3% for R2) than R3 (55%), suggesting the effects of feeding strategy on EBPR. Wang et al. also found that a 70% removal of total inorganic nitrogen (TIN) was achieved by granular sludge in a continuously plug–flow bioreactor fed with real domestic wastewater, meeting the discharge limit of TIN < 10 mg/L in. These studies collectively demonstrate that aerobic granules are highly effective in removing nitrogen and phosphorus from domestic wastewater, side streams, and nitrogen-rich wastewater. However, achieving optimal results depends on the optimisation of both the process and the operation for the specific wastewater with different reactor and process configurations such as the feeding strategy (Mady et al.) and the nitrate recycling location. In addition, a rapid conversion of heterotrophic granules into nitrifying granules [4] has practical implications for quick-start-up nitrifying granules to treat nitrogen-rich wastewater.

In addition to nutrients, it is crucial to address other pollutants since municipal wastewater treatment plants often receive a portion of industrial wastewater when necessary. Ortega et al. investigated the effects of increased particulate COD (pCOD) on the aerobic granular sludge process in a full-scale Nereda® plant receiving (30%) raw industrial wastewater mainly from slaughterhouses with an increased pCOD load from 0.5 to 1.3 g COD/L. Remarkably, no deterioration in granule properties or reactor performance, such as nutrient removal, was observed. This underscores the robustness of the granular sludge process in handling more challenging wastewater types.

Aerobic granules are also effective in treating industrial wastewater. This Special Issue includes a study on berberine wastewater from the pharmaceutical industry. By developing a hybrid anaerobic baffled reactor–aerobic granules process (ABR–AGS) system fed with wastewater containing a berberine concentration of 122 mg/L, Wang et al. achieved 92.2% and 94.8% overall removals of berberine and COD, respectively. Due to their compact structure and large size, aerobic granules are capable of withstanding toxic and inhibitory substances such as berberine, making them a more promising solution for treating harsher wastewater compared to suspended sludge. Experimental evidence from Tay et al. [5] demonstrated that unlike activated sludge reactors, aerobic granules quickly adapted and efficiently reduced influent phenol concentrations from 600 mg/L to 0.3 mg/L within three days after a substrate change from acetate to phenol. When the algal–bacterial aerobic granular sludge was used as biosorbent for treating wastewater containing heavy metals such as 2 mg/L Cr(VI) at a 1% salinity, the removals of total inorganic nitrogen (TIN) and total phosphorus (TP) were noticeably impacted, and they were further decreased with the co-existence of 2 mg/L Cr(VI), but the Cr(VI) removal efficiency was only influenced slightly by salinity exposure. Nitrifying bacteria are sensitive to unfavourable environmental conditions such as high salinity and high heavy-metal concentrations. Although granules' structure can protect bacteria to a certain extent, achieving the removal of both nutrients and heavy metals at a high salinity is challenging, but based on this study, using waste granular sludge as a biosorbent for Cr(VI) removal at high salinity seems viable.

For various types of wastewater mentioned above, such as that with a low COD/N ratio, side streams with high nutrient concentrations, high-pCOD wastewater, and pharmaceutical wastewater containing berberine, the aerobic granular sludge process generally demonstrates high robustness and good performance, implying its versatility for treating various types of wastewater. However, optimisation has to be conducted for each type of wastewater to ensure a good performance, and the treatment mechanism needs to be understood for better control and operation of aerobic granular process.

Long-Term Stability and Relevant Performance of Aerobic Granules in Various Treatment Processes

The long-term stability of aerobic granular sludge and reactor performance is another important factor affecting the adoption of this technology and its practical implementation. In this Special Issue, three types of reactor operation were studied: conventional SBRs, continuously plug–flow reactors for process intensification, and continuous-flow granular membrane bioreactors (GMBRs). Two pilot-scale plug–flow reactor (PFR) systems, based on a modified Ludzack–Ettinger (MLE) configuration with nitrate recycling on continuous-flow aerobic granulation fed with real domestic wastewater, were used to study the effects of nitrate recycle location on granulation. The results showed that the change in nitrate recycle location can be advantageous to drive sludge densification without a radical washout of the sludge, and it had no effects on the chemical oxygen demand (COD) and nitrogen removal efficiencies. Thus, it could be further developed as an alternative to hydrocyclones for full-scale, greenfield, continuous sludge densification applications. In conventional lab-scale SBRs, duplicate reactor operation with a low COD/N ratio (4/1) showed a slight difference for granulation. Additionally, the SVI_{30}, granule size, and size distributions of sludge in two reactors were different. However, the granulation and reactor performance in terms of nutrient removal were generally reproducible. This study validated the reproducibility of aerobic graduation and relevant treatment performance in duplicates, but meanwhile, it suggests that slight differences between different reactors and granule properties are unavoidable. This result is meaningful because the vast majority of studies on aerobic granules had no duplicate reactor operation and the comparison between different conditions, especially when the differences are not significant, could be better interpreted. Since there is typically no deliberate sludge wasting or control of biomass concentration in lab-scale reactors as in full-scale wastewater treatment plants, the biomass increase due to the granulation and biomass accumulation results in a reduced F/M ratio, which is believed to be critical for the long-term stability of aerobic granules [6]. Controlling F/M ratios in a certain range would thus benefit long-term stability. Liu et al. observed a serious calcification of partial nitrifying granules during the long-term operation with ash contents varying from 50% to 85% at stable state at 21 °C and 32 °C, fed with wastewater with high hardness (Ca^{2+} concentration > 120 mg/L). The precipitation and accumulation of hydroxyapatite and calcium carbonate in granules resulted in a significantly reduced specific nitrogen oxidation rate, and the higher temperature exacerbated this phenomenon. It was concluded that high mineral content in granules led to the instability of nitrifying granules at a higher temperature in the short or long term. The effects of water hardness on granule stability are not well investigated; thus, this study provided insights for further investigation of this aspect in the future, especially when considering adopting aerobic granule technology in areas with high water hardness. It needs to be pointed out that water hardness is not the only factor that causes the calcification of aerobic granules. The high mineral content in aerobic granules has been reported under other conditions as well. In this Special Issue, An et al. found that in GMBR without sludge wasting, inoculated nitrifying granules disintegrated and re-established over a 300-day operation, suggesting that membrane module and indefinite SRT did not affect granulation although there was granule disintegration first. However, the ash content in granules was as high as 64% after a 305-day operation with only the top 60–80 μm layer of these nitrifying granules being active, which is similar to a biofilm with an active layer on the surface of the inert carrier media. The rate of membrane fouling did not change in this GMBR, thus defeating the original purpose of GMBR development. It is noteworthy that SRT in this GMBR was indefinite because there was no sludge wasting and the reactor was operated in continuous mode. Other conditions of GMBR such as controlling SRT need further investigation to fully explore the potential of GMBR for the alleviation of membrane fouling. Generally, retaining granules in reactors for a long period, i.e., long SRT, could cause granule calcification, leading to the increase in ash content for reduced microbial activity [7], possible granule disintegration, and reactor operation instability. Thus, controlling the SRT of granules (sometimes F/M ratios) could be an effective strategy to minimise the increase in ash content and maintain the long-term stability of granules in terms of structure as well as microbial activity.

Digestibility of Waste Aerobic Granules

Just like conventional activated sludge-based plants, anaerobic digestion is a common approach to treating waste sludge in aerobic-granule-based WWTPs. The physical and chemical characteristics of granular sludge are different from those of activated sludge, which could lead to different digestibility and biogas production potential. A review paper by Zaghloul et al. [8] discussed the biological and chemical characteristics of waste AGSs and their digestibility. In addition, the comparison between aerobic granules, activated sludge, and primary sludge in terms of digestibility was conducted. It concluded that the AGS morphology and structural polymers slowed down the biomethane production rates, but the overall yield was on par with that of activated sludge. So far, the available literature on the digestibility of waste AGS is still scarce compared to WAS. Thus, further research, especially on waste AGS from full-scale plants, was suggested. In addition, aerobic granules could be used for various types of wastewater treatment in different types of reactors, from which characteristics of waste AGS and digestibility should be investigated to draw a solid conclusion.

In summary, this Special Issue addresses the key aspects of aerobic granular sludge in environmental protection. It encompasses a variety of topics including different types of wastewater characterised by diverse influent qualities, particularly varying nutrient levels. The Special Issue also explores different operation modes such as sequential-batch and continuous-flow operation modes, with different process configurations such as SBRs, GMBRs, and continuous plug–flow reactors. Significantly, the studies extend beyond synthetic wastewater to include real domestic wastewater, municipal wastewater mixed with industrial water (such as slaughterhouse water), and actual industrial wastewater like berberine wastewater. These studies, conducted in lab-scale, pilot-scale, and full-scale reactors, deepen our understanding of the practical application of aerobic granules in real-world scenarios. They demonstrate the robustness of aerobic granules in treating a wide array of wastewater types, highlighting their promising potential for widespread application. Furthermore, some key factors in different process configurations with different wastewater were investigated to understand the long-term stability of both granule structure and reactor performance. SRT is one of the most crucial operating parameters which might affect granule calcification and disintegration. Lastly, a review paper discussed an interesting finding of comparable digestibility and biogas yields between aerobic granules and activated sludge, although the methane production rate of granules could be lower due to different physical and chemical properties. This area, still under-researched, calls for further studies. Moreover, many papers in this Special Issue utilised 16S RNA gene sequencing to analyse the microbial community structure of aerobic granules under different treatment conditions. However, a detailed correlation between the microbial community structure and operating conditions or granule properties was not established. Future research in this area could provide valuable insights for improved process control and operation. Overall, this Special Issue serves as a microcosm of the current research on aerobic granules, offering a quick glimpse into the state of the art in aerobic granules for environmental protection.

Conflicts of Interest: The author declares no conflicts of interest.

List of Contributions:

1. Zhang, H.; Liu, Y.Q.; Mao, S.; Steinberg, C.E.W.; Duan, W.; Chen, F. Reproducibility of aerobic granules in treating Low-strength and low-C/N-ratio wastewater and associated microbial community structure. *Processes* **2022**, *10*, 444.
2. Liu, Y.Q.; Cinquepalmi S. Hydroxyapatite precipitation and accumulation in granules and its effects on activity and stability of partial nitrifying granules at moderate and high temperatures. *Processes* **2021**, *9*, 1710.
3. Morgan, G.; Hamza, R.A. Cultivation of Nitrifying and Nitrifying-Denitrifying Aerobic Granular Sludge for Sidestream Treatment of Anaerobically Digested Sludge Centrate. *Processes* **2022**, *10*, 1687.
4. An, Z.; Zhang, X.; Bott, C.B.; Wang, Z.W. Long-term stability of nitrifying granules in a membrane bioreactor without hydraulic selection pressure. *Processes* **2021**, *9*, 1024. https://doi.org/10.3390/pr9061024.
5. Iorhemen, O.T.; Ukaigwe, S.; Dang, H.; Liu, Y. Phosphorus removal from aerobic granular sludge: Proliferation of polyphosphate-accumulating organisms (PAOs) under different feeding strategies. *Processes* **2022**, *10*, 1339.
6. Wang, J.F.; An, Z.H.; Zhang, X.Y.; Angelotti, B.; Brooks, M.; Wang, Z.W. Effects of Nitrate Recycle on the Sludge Densification in Plug-Flow Bioreactors Fed with Real Domestic Wastewater. *Processes* **2023**, *11*, 1876.
7. Ortega, S.T.; Pronk, M.; de Kreuk, M.K. Effect of an increased particulate cod load on the aerobic granular sludge process: A full scale study. *Processes* **2021**, *9*, 1472. https://doi.org/10.3390/pr9081472.
8. Wang, Y.; Liu, Y.; Li, J.; Ma, R.; Zeng, P.; Ng, C.A.; Liu, F. The dynamic shift of bacterial communities in hybrid anaerobic baffled reactor (ABR)—Aerobic granules process for Berberine pharmaceutical wastewater treatment. *Processes* **2022**, *10*, 2506. https://doi.org/10.3390/pr10122506.
9. Van Nguyen, B.; Yang, X.; Hirayama, S.; Wang, J.; Zhao, Z.; Lei, Z. Effect of salinity on cr(Vi) bioremediation by algal-bacterial aerobic granular sludge treating synthetic wastewater. *Processes* **2021**, *9*, 1400.
10. Zaghloul, M.S.; Halbas, A.M.; Hamza, R.A.; Elbeshbishy, E. Review on Digestibility of Aerobic Granular Sludge. *Processes* **2023**, *11*, 326.
11. Mady, E.; Oleszkiewicz, J.; Yuan, Q. Feasibility Study of Applying Anaerobic Step-Feeding Mode for the Treatment of High-Strength Wastewater in Granular Sequencing Batch Reactors (GSBRs). *Processes* **2023**, *11*, 75.

References

1. Mishima, K.; Nakamura, M. Self-immobilization of aerobic activated sludge—A pilot study of the Aerobic Upflow Sludge Blanket Process in municipal sewage treatment. *Water Sci. Technol.* **1991**, *23*, 981–990. [CrossRef]
2. Morgenroth, E.; Sherden, T.; Van Loosdrecht, M.C.M.; Heijnen, J.J.; Wilderer, P.A. Aerobic granular sludge in a sequencing batch reactor. *Wat. Res.* **1997**, *31*, 3191. [CrossRef]
3. Liu, Y.Q.; Cinquepalmi, S. Hydroxyapatite precipitation and accumulation in granules and its effects on activity and stability of partial nitrifying granules at moderate and high temperatures. *Processes* **2021**, *9*, 1710. [CrossRef]
4. Liu, Y.Q.; Tay, J.H. Fast formation of aerobic granules by combining strong hydraulic selection pressure with overstressed organic loading rate. *Water Res.* **2015**, *80*, 256–266. [CrossRef] [PubMed]
5. Tay, S.T.L.; Moy, B.Y.P.; Maszenan, A.M.; Tay, J.H. Comparing activated sludge and aerobic granules as microbial inocula for phenol biodegradation. *Appl. Microbiol. Biotechnol.* **2005**, *67*, 708–713. [CrossRef] [PubMed]
6. Franca, R.D.G.; Pinheiro, H.M.; van Loosdrecht, M.C.M.; Lourenço, N.D. Stability of aerobic granules during long-term bioreactor operation. *Biotech. Adv.* **2018**, *36*, 228.
7. Yuan, S.; Gao, M.; Zhu, F.; Afzal, M.Z.; Wang, Y.-K.; Xu, H.; Wang, M.; Wang, S.-G.; Wang, X.-H. Disintegration of aerobic granules during prolonged operation. *Water Res. Technol.* **2017**, *3*, 757. [CrossRef]
8. Zaghloul, M.S.; Halbas, A.M.; Hamza, R.A.; Elbeshbishy, E. Review on Digestibility of Aerobic Granular Sludge. *Processes* **2023**, *11*, 326. [CrossRef]