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The Aerobic Granules Process for Wastewater Treatment: From Theory to Engineering

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Abstract: Aerobic granules are small, dense aggregates of microbial cells that form naturally in aerobic wastewater treatment systems. They are characterized by their spherical shape, strong structural integrity, and ability to rapidly settle. These granules are formed through a self-immobilization process where different microbial species coalesce to degrade organic and inorganic compounds in wastewater. This study summarizes the development of aerobic granulation technology in wastewater treatment and the mechanism of aerobic granules' formation, analyzes the characteristics and the factors affecting the aerobic granules' formation, and presents practical engineering examples of its application from pilot-scale to full-scale operation.

Keywords: aerobic granular reactor; SBR; CFR; full-scale reactor; mechanism

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1. Introduction

Successful wastewater treatment relies on choosing microorganisms with the right metabolic capabilities and effectively separating them from the treated effluent. Significant research efforts have been directed towards minimizing the settling time of activated sludge. This is achieved by forming dense flocs or employing biofilm reactors. Biogranules, a condensed form of biofilm created through self-immobilization, represent a noteworthy innovation in this field [1]. They can be classified as aerobic or anaerobic granulation. These granules are compact conglomerates of diverse bacterial species, with each gram of biomass harboring millions of organisms.

The development of anaerobic granules is a well-researched area, particularly noted for its application in the upflow anaerobic sludge blanket (UASB) reactor. This technology, known as anaerobic granulation, has been implemented in numerous wastewater treatment facilities [2–5]. In reactors that utilize granular sludge, these dense anaerobic granules settle quickly. This swift settling significantly cuts down the time needed to separate the treated water from the biomass. The distinctive characteristics of anaerobic granules have captivated researchers, motivating them to focus on the development of aerobic granulation technology. This shift aims to expand the application spectrum of granulation techniques, potentially enhancing their utility in a wider range of scenarios.

Aerobic granules are small, dense aggregates of microbial cells that form naturally in aerobic wastewater treatment systems. They are characterized by their spherical shape, strong structural integrity, and ability to rapidly settle. These granules are formed through a self-immobilization process where different microbial species coalesce to degrade organic and inorganic compounds in wastewater. This paper aims to comprehensively summarize the development process of aerobic granules. It will delve into the formation mechanism, explore the distinct characteristics, and examine the various factors influencing the formation of aerobic granules. The paper will culminate with a discussion on the application of these granules, focusing particularly on their implementation in pilot-scale and full-scale settings.

2. The Development of Aerobic Granular Technology

2.1. Aerobic Granulation in Sequencing Batch Reactors

Aerobic granular sludge technology began to emerge in the early 1990s [6]. Toward the late 1990s, there was a notable surge in research focused on the basic principles and practical applications of aerobic granulation. This led to significant advancements in the understanding of how storage polymers contribute to the development of aerobic granules, facilitating their growth in sequencing batch reactors without the need for carrier materials [7–9].

Aerobic granular sludge technology developed from the early 1990s [6]. Research interest in the fundamental concepts and real-world uses of aerobic granulation intensified towards the late 1990s. Investigations into the creation of storage polymers have paved the way for progress in the growth of aerobic granules in sequencing batch reactors (SBR) without the necessity for carrier materials [7–9]. The sequencing batch reactor (SBR), originating in the United States in the late 1960s, gained widespread adoption throughout the 1980s and 1990s [10]. In contrast to the operation of a suspended sludge SBR, an aerobic granular sludge SBR does not include an idling phase in its operation.

The initial observation of aerobic granule formation in SBR was made by Morgenroth et al. [7]. They operated the SBR with a notably brief sedimentation and draw phase, which led to the removal of biomass with a low settling ability. After 40 days of this operation, granules became the primary form of biomass in their reactor. Then, a large number of papers on aerobic granular sludge cultivation in the laboratory were published.

In 2003, the Ede wastewater treatment facility in The Netherlands became the site of the world's inaugural pilot-scale operation of aerobic granular sludge technology [11]. In 2004, the first international seminar on aerobic granular sludge was held at the Technical University of Munich in Germany, where the concept of aerobic granular sludge was introduced [12]. In 2005, the Dutch company DHV developed the Nereda process, which was successfully applied for the first time in a wastewater treatment plant [13]. In 2014, the aerobic granular sludge technology was successfully applied in Yancang WWTP in the Zhejiang province of China [13]. To date, aerobic granular sludge technology has been applied in over 100 WWPTs worldwide by Nereda® (Royal haskoningDHV, Amersfoort, The Nertherland) [14]. The development process of aerobic granulation technology in SBR is shown in Figure 1.

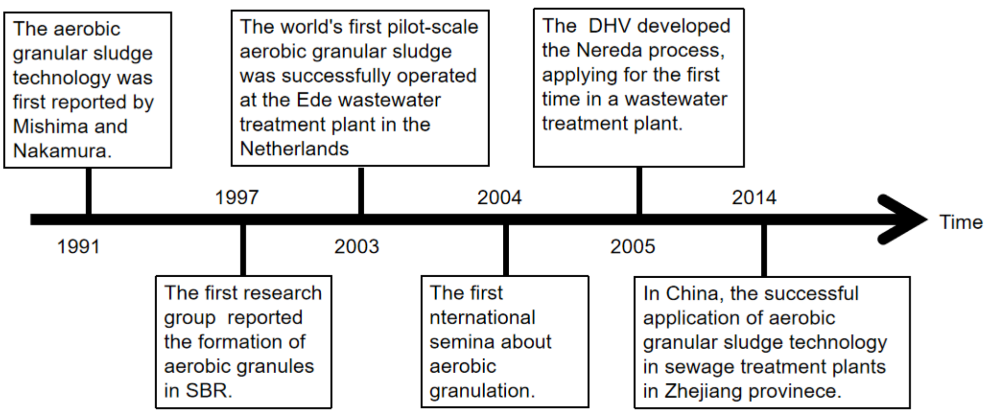

Figure 1. The development of aerobic granulation technology in SBR.

2.2. Aerobic Granulation in Continuous-Flow Reactors

Reports have also indicated the occurrence of aerobic granulation in continuous-flow systems, specifically in an aerobic upflow sludge blanket (AUSB) reactor [6,15], a fluidized-bed reactor [16], an airlift biofilm reactor [17], a flow baffled reactor [18] and an upflow membrane-aerated biofilm reactor [19].

In the AUSB reactor, the granules are formed with low shear forces. This process is driven by vertical stresses from the upward flow of liquid and horizontal stresses caused by agitation at a rate of 1–6 rpm. Granule formation typically occurs within approximately 5 days [15], or up to three weeks [6], after initiation. However, the complexity of the AUSB reactor system, along with its specific requirements for pure oxygen (i.e., 100% O_2) and a dissolved oxygen tank, may hinder its broader adoption.

In two-phase fluidized-bed reactors, the formation of nitrifying granules is likely due to the relatively low shear forces and self-aggregation. Additionally, nitrifying granules can form in airlift reactors, originating from fragments of broken biofilm. The depletion of oxygen within the nitrifying biofilm leads to its breakup, and the resulting dense biofilm fragments remain in the reactor. Generally, some carrier material is necessary to ensure system stability.

Unlike traditional SBR aerobic granular sludge, continuous-flow reactors do not have a static sedimentation process [20,21]. Therefore, an additional sludge–water separation device is needed in the reactor for sludge–water separation. Based on the different sludge–water separation mechanisms, continuous-flow aerobic granular sludge reactors can be classified into the gravity sedimentation separation type, three-phase separation type, and filtration separation type. The development of aerobic granulation technology in continuous-flow reactors is shown in Figure 2.

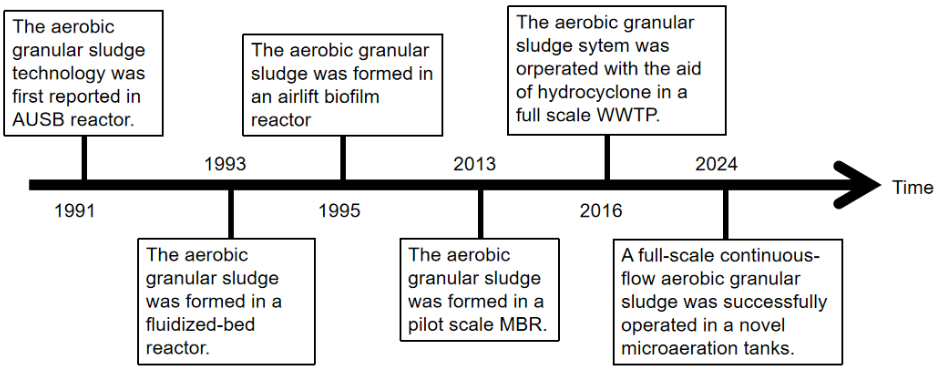

Figure 2. The development of aerobic granulation technology in continuous-flow reactors.

3. Mechanism of Aerobic Granulation

The process underlying microbial aerobic granulation remains a subject of significant debate due to the intricate nature of aerobic granulation. According to the hypothesis summarized in Table 1, aerobic granulation starts with the aggregation of microbial cells. Fungi, using glucose as their carbon source, secrete H^+ ions to lower the pH, creating an environment conducive to their growth while suppressing competing bacteria [22]. Filamentous fungi grow from spores to mycelia under shear forces, serving both as an attraction nucleus for other microorganisms and as a structural framework. Some bacteria attach to inorganic precipitates within the granule's core, with these precipitates acting as a nucleus for growth and reproduction. Other bacteria use cations like Ca^{2+} and Mg^{2+} to promote aggregation by reducing electrostatic repulsion and enhancing van der Waals forces. Shear forces in the granulation process stimulate the secretion of EPS, binding organisms within the granule. The increased cell surface hydrophobicity and high hydraulic stress, combined with the limited settling time, encourage microorganisms to adhere to aggregates, forming aerobic granules.

Table 1. The proposed mechanism of aerobic granulation.

Year	Researcher	The Proposed Mechanism	Reference
1999	Beun et al.	(1) Aerobic granulation started with fungi. (2) Fungi easily formed pellets. When the pellets grew up and lysed, (3) the pellets broke apart, and only the adequately dense colonies were able to settle successfully. (4) As time progressed, these colonies expanded and transformed into newly formed granules.	[8]
2001	Tay et al.	With the condition of sequential operation, aerobic granules originate from seed sludge, progress to compact aggregates, then develop into granular sludge, and ultimately evolve into mature granules.	[23]
2002	Liu and Tay	(1) Cells moving randomly and colliding; (2) cells moving randomly and colliding; (3) continued irreversible clustering and expansion within the matrix of extracellular polymeric substances (EPSs); and (4) the formation of shape and structure, influenced by shear forces.	[24]
2004	Liu et al.	Hydraulic pressure is a key factor in the formation of biogranules, whereas the hydrophobicity of cells markedly contributes to the development of granules. Furthermore, the formation of aerobic granules results from the combined efforts of different functional groups and their interactions with the ambient environment.	[25]
2010	Barr et al.	(1) A single microbial colony can gradually expand to form compact and smooth granules. (2) The aggregation of multiple independent microbial colonies can lead to the formation of relatively loose granules.	[26]
2015	Wu et al.	Under continuous flow conditions, the formation of aerobic granules is critically dependent on two key factors: a high organic loading rate and intense selection pressure.	[27]
2022	Edward et al.	(1) Selection of microorganisms, (2) targeted substrate utilization, (3) enhancing substrate transport into the biofilm, (4) specific feeding strategies, (5) substrates that either form or do not form particles, (6) breakdown of granules.	[28]

Granules initially form from small microbial clusters that grow via further aggregation. These dense granules settle quickly due to selection pressure in the reactor, favoring fast-settling biomass and eliminating slower settlers. The oxygen and nutrient gradients within granules create different aerobic and anaerobic zones, allowing diverse microbial communities to contribute to the granules' structure and function. Granules mature and grow, balancing microbial proliferation, EPS production, and shear forces that might cause breakup. The reactor's operation, including the feed composition, aeration, and cycle management, is vital for sustaining granulation.

4. Characteristics of Aerobic Granules

Aerobic granular sludge is distinct from flocculent sludge. Its characteristics are outlined as follows [29–40]:

It possesses a spherical and uniform shape with a distinct, smooth exterior;
It exhibits a tightly packed and robust microbial constitution;
It is sufficiently large to be visible as individual entities in the mixed liquor during both the mixing and settling phases.
Sludge has a high biomass retention capability caused by its large size and fast settling velocity.
It is capable of enduring high organic loading rates.
It is resilient against the toxicity substances present in wastewater.

Table 2 provides comprehensive details of aerobic granular sludge.

Table 2. The characteristic data about aerobic granular sludge.

Reactor	Diameter (mm)	SVI (mL g^{-1})	MLSS (g L^{-1})	Specific Gravity	Settling Velocity (m h^{-1})	Substrate	Reference
SBR	0.6–1.4	30–40	5	1.021	22–60	Synthetic wastewater with sodium acetate as the main carbon source, COD: 500 mg L^{-1}	[41]
SBAR	0.3–3		7–10	-	-	Influence synthetic wastewater with an acetate concentration of 18.3 Cmmol L^{-1}	[42]
SBR	0.3–0.5	80–100	-	-	-	Acetate	[9]
SBR	2	172	2	1.0038	-	Phenol	[43]
SBR	2.8	73	7.9	1.0068	-	Phenol with Ca^{2+}	[43]
CFR system with baffled bubble column	0.2–2	33.5	2.8–5.8	-	-	Synthetic wastewater with sodium acetate as the main carbon source, COD:1500 mg L^{-1}	[44]
CFR system with multiple serial chambers	0.13	43	3.0	-	-	Municipal (30%) and industrial (70%) wastewater	[45]
CFR system with MBR	0.1–1.0	25–40	10	-	15–25	Synthetic wastewater with glucose as the main carbon source, COD: 100–300 mg L^{-1}	[46]

The reactor type, feeding substrate, and operation conditions significantly influence the characteristics of aerobic granules. Specifically, the choice of reactor, such as the Sequencing Batch Reactor (SBR) or Continuous-Flow Reactor (CFR), affects the formation, size, and density of the granules due to differences in the flow dynamics and mixing patterns [40–44]. The nature of the feeding substrate, including its composition and concentration, impacts the microbial community structure within the granules, as well as their metabolic capabilities and pollutant removal efficiency. For instance, high-strength organic substrates can lead to larger and denser granules with a diverse microbial community. Lastly, operation conditions such as hydraulic retention time (HRT), sludge retention time (SRT), aeration intensity, and temperature play crucial roles in determining the physical strength, settling velocity, and overall stability of aerobic granules. Optimizing these parameters is essential for achieving efficient wastewater treatment and enhancing the robustness and resilience of aerobic granular sludge systems.

5. Structure and Strength of Aerobic Granule

Aerobic granules, spherical biofilms encapsulated within an extracellular polymer (ECP) matrix, exhibit a complex, layered structure that is critical for their functionality. Advanced imaging techniques, such as Confocal Laser-Scanning Microscopy (CLSM), have been instrumental in delineating these layers, with each serving distinct roles in the granules' overall efficacy and stability [47–53]. Researchers have uncovered a layered structure within these granules [9]. In granules fed with acetate, the outermost layer is composed of viable cells, lysed cells, non-degradable cellular remnants, and solids from the influent, constituting a layer that is 0.5–5 μm in thickness. Following this, the second layer is made up of distinct aggregates enveloped in well-defined polymeric matrices. These aggregates form roughly spherical microcolonies, varying in size from 5 to 50 μm. The third layer is formed by embedding these numerous microcolonies within the extracellular polymer [49].

Notably, a layer of deceased microbial cells is situated about 800–1000 μm beneath the granules' surface, and the anaerobic bacteria *Bacteroides* spp. have been detected at a depth of 800–900 μm from the surface. The production of polysaccharides in these granules reaches its peak at approximately 400 μm below the surface [50]. The granules feature channels and pores extending up to 900 μm beneath the surface, with the highest porosity observed at depths ranging from 300 to 500 μm. These channels and pores play a crucial role in facilitating the movement of oxygen and nutrients into the granules, as well as the exit of metabolites. The arrangement of pores and channel layers within the granules affects the distribution of the active biomass, which varies across granules of different diameters. The porosity of these granules is critical to their strength and stability, enabling them to withstand the mechanical stresses encountered in wastewater treatment processes.

The relationship between the structure and strength of aerobic granules is thus deeply intertwined. The granules' layered structure, with its distinct microbial compositions and functions, underpins their strength, both in terms of physical integrity and metabolic capacity. This structural strength is vital for the granules' resilience against physical shearing forces in reactors and their ability to maintain high rates of pollutant degradation. Moreover, the size and arrangement of pores and channels within the granules directly impact their effectiveness in nutrient and oxygen diffusion, further influencing the granules' strength and stability.

6. Diversity of Aerobic Granules

To better understand the process of aerobic granulation and enhance the design and operational efficiency of aerobic granulation systems, extensive studies have delved into the microbial diversity within aerobic granules, employing advanced molecular biotechnology methods [43,54–59]. These investigations have unveiled a broad spectrum of bacteria, demonstrating the presence of heterotrophic bacteria, nitrifying and denitrifying bacteria, phosphorus-accumulating bacteria, and glycogen-accumulating bacteria, cultivated under a range of conditions and using various culture media [23,46,54,55,58,60–62]. Further exploration into the growth patterns of these microorganisms reveals a complex interplay between reactor configurations, substrate types, and operational parameters. For instance, the presence of heterotrophic bacteria is significantly influenced by the organic carbon source and its availability, which, in turn, affects the granule formation by facilitating biomass aggregation and structure stability. Nitrifying and denitrifying bacteria, on the other hand, are crucial for nitrogen removal and are found to proliferate under specific aeration and substrate concentration conditions, contributing to the structural integrity and functional diversity of the granules. Phosphorus-accumulating organisms play a pivotal role in biological phosphorus removal and are encouraged by alternating anaerobic and aerobic conditions, which also affect the granule's density and settling properties. Additionally, the role of glycogen-accumulating organisms in managing the internal carbon flux within granules under fluctuating feeding conditions highlights the adaptive mechanisms of microbial communities in aerobic granules.

The interdependence between these microbial communities under varied environmental and operational conditions underscores the complexity of aerobic granulation. Such diversity not only contributes to the robustness and resilience of granular sludge systems but also offers insights into the optimal conditions for enhanced pollutant removal efficiency. This expanded understanding emphasizes the need for tailored approaches to reactor design and operation to harness the full potential of microbial diversity in aerobic granulation processes.

7. Factors Affecting the Formation and Structure of Aerobic Granule

7.1. Shear Force

In SBR, the shear force is primarily determined by the upflow air velocity. Studies have suggested that a higher shear force favors the creation of aerobic granules [8,15,22]. For instance, Tay et al. found that a superficial air upflow velocity of 0.8 cm s^{-1} led to the formation of only loose flocs in an SBR, while a higher velocity of 2.5 cm s^{-1} facilitated the formation of well-shaped granules. This observation was also noted by Beun et al. [63], reinforcing the idea. Additionally, there is a proportional relationship between granule density and strength and the applied shear force. These insights reveal that shear force plays a vital role in both the development of aerobic granulation and in shaping the granules' structure.

Moreover, Tay et al. [22] reported a close association between the production of cellular polysaccharides and shear force, which further contributes to the stability of aerobic granules. This suggests that the shear force not only encourages the formation of aerobic granules but also stimulates the production of cellular polysaccharides, thereby playing an essential role in their development.

In addition, the upflow velocity, indicated by the shear force, affects the hydrodynamic conditions within the reactor, influencing the collision and adhesion rates of microbial cells. Optimal upflow velocities can enhance the collision rate between microbial cells and flocs, promoting granulation. However, too high or too low a velocity can disrupt this balance, affecting the overall size and stability of the granules.

7.2. Settling Time

The settling time plays a pivotal role as a form of hydraulic selection pressure on the microbial populations during the process of aerobic granulation. A reduced settling time is crucial for two main reasons: firstly, it can eliminate slowly settling biomass while preserving granules that settle quickly, and secondly, it can guarantee the biological systems operate both efficiently and cost-effectively. Studies have demonstrated that aerobic granules became the predominant form in an SBR when the system was managed with a settling time of only 5 min [64]. Extending the settling time beyond 5 min resulted in a mixed culture of aerobic granules and free-floating sludge. In practice, the settling time is typically maintained between 1 and 4 min [7,8,65]. Thus, identifying the ideal settling time is essential for successful aerobic granulation.

7.3. Organic Loading Rate

While a high Organic Loading Rate (OLR) is beneficial to the formation of anaerobic granules in UASB systems, it tends to be detrimental to aerobic granulation due to the excessive proliferation of suspended forms. Aerobic granules can successfully form within a range of COD loading rates, from 0.42 to 15 kg COD $m^{-3} \cdot d^{-1}$ [62–64]. Although OLR does not significantly impact the initial formation of aerobic granules, it is closely linked to their physical characteristics.

Toh et al. [66] noted that the average diameter of aerobic granules tends to expand as the OLR increases [67–69]. Nevertheless, characteristics such as the granules' roundness, dry biomass density, specific gravity, and Sludge Volume Index (SVI) do not show a significant relationship with the OLR. On the other hand, the structural integrity of aerobic granules weakens with an increase in OLR. This reduction in physical strength is linked to the rapid growth in biomass caused by higher OLRs, which adversely affects the three-dimensional structure of the microbial aggregates [70].

7.4. Substrate Composition

A diverse range of substrates, including glucose, acetate, ethanol, phenol, yeast extract, particulate organic-matter-rich wastewater, and berberine wastewater, have been effectively utilized to cultivate aerobic granules [64–69]. Additionally, granules with specific nitrification and phosphorus accumulation abilities have been engineered [70–74]. It is crucial to recognize that the choice of substrate can profoundly affect the microbial diversity, microstructure, and elemental makeup of mature aerobic granules [71].

For example, aerobic granules formed using acetate as a substrate tend to have a dense microstructure with predominantly radially arranged, rod-like bacteria. Conversely, granules cultivated with glucose predominantly contain cocci-type bacteria internally, with a surface layer composed of both filamentous and rod bacteria. These differences highlight the significant role that substrate type plays in determining the physical and biological properties of aerobic granules.

7.5. EPS

Extracellular Polymeric Substances (EPS), secreted by microorganisms, are intricate combinations of proteins, polysaccharides, humic substances, and nucleic acids. These substances are instrumental in aerobic granulation.

Initially, EPS are pivotal to biofilm formation and development, serving as the foundational element of aerobic granules. They facilitate the adhesion of microbial cells to one another and to various surfaces, thereby fostering the creation of dense microbial communities that are essential for effective wastewater treatment.

As granulation progresses, EPS form a structural matrix that encapsulates the microbial cells within the granules. This matrix not only bestows shape and structural integrity upon the granules but also supports the bacterial cells' aggregation, enhancing the granules' mechanical stability.

Furthermore, the EPS matrix acts as a protective barrier for the microbial cells against environmental stressors such as toxic substances, pH fluctuations, and shear forces, ensuring their viability and functionality within the wastewater treatment process.

EPS's ability to bind nutrients, metals, and other substances plays a critical role in their retention within the granules and facilitates exchange within the microbial community. This trait is vital for the microorganisms' metabolic activities, improving the wastewater treatment's overall efficiency. Additionally, during periods of scarcity, the degradation of EPS components like proteins, polysaccharides, and humic compounds serves as a carbon source, increasing cell surface hydrophobicity and enhancing granule stability.

Moreover, EPS act as conduits for cell-to-cell communication within granules, orchestrating metabolic activities and enabling the microbial community to adapt to environmental changes.

7.6. Hydraulic Retention Time

Hydraulic Retention Time (HRT) is a critical factor in the functioning of SBR, associated with both the cycle duration and the volume exchange rate of the SBR. Defined by the ratio between the volume of effluent that is released and the SBR's operational volume, HRT significantly influences the granulation process as a form of hydraulic selection pressure, deterring the proliferation of dispersed sludge. Selecting a suitable HRT is essential for encouraging the development of granules [72–74].

An HRT that is too short could lead to significant sludge loss without adequate compensation, negatively impacting the system's efficiency. On the other hand, an excessively long HRT could result in bioflocs becoming dominant in the system, which might not be desirable depending on the specific goals of the treatment process. It has been observed that the stabilization of seeded aerobic granules is achievable with an HRT ranging from 2 to 12 h [73]. Furthermore, HRTs of 12 and 24 h are particularly beneficial for nitrifying bacteria, indicating that the optimal HRT can vary depending on the specific microbial community or process goals within the SBR [74].

The strategy of controlling HRT within a moderate range is also applied to CFRs, given their incorporation into an internal settling zone [75–78].

7.7. Dissolved Oxygen

The Dissolved Oxygen (DO) concentration is indeed a vital parameter in managing aerobic wastewater treatment systems. However, when it comes to the formation of aerobic granules, the DO concentration does not play a decisive role. Research has shown that aerobic granules can successfully form even at relatively low DO concentrations, specifically in the range of 0.7–1.0 mg L^{-1} in an SBR [9]. Additionally, aerobic granules have been successfully developed at DO concentrations exceeding 2.0 mg L^{-1}. This indicates that aerobic granules can form and sustain under a broad range of DO concentrations, making them adaptable to various operating conditions in aerobic wastewater treatment processes [43,69].

7.8. Aerobic Starvation

In a typical SBR cycle, there are two distinct phases: the degradation phase and the aerobic starvation phase. During the degradation phase, the substrate is progressively depleted to a minimal level. This phase is succeeded by the aerobic starvation period, during which the microorganisms in the SBR lack access to any external substrate.

Consequently, these microorganisms experience periodic starvation. It has been proposed that this intermittent starvation could induce microbial adhesion and aggregation, possibly through its impact on cell hydrophobicity [79,80]. This concept suggests that the stress of intermittent starvation could alter the surface properties of the cells, making them more likely to adhere to each other and form aggregates. Nonetheless, it is critical to acknowledge that, despite thorough investigations into the link between starvation and cell hydrophobicity, the particular notion that it initiates microbial adhesion and aggregation within SBR systems lacks direct validation through experimental proof. Further research is needed to validate this hypothesis and fully understand the mechanisms at play.

7.9. Trace Elements

Trace elements, particularly calcium and iron, play a significant role in enhancing aerobic granulation. The study by Jiang et al. demonstrated that adequate amounts of calcium can significantly expedite the formation of aerobic granules, reducing the required time by about 50% [70]. These calcium-augmented aerobic granules not only settle better but also exhibit improved strength characteristics and have an increased polysaccharide content. Ca^{2+} are thought to act as a bridge that promotes bacterial aggregation. They achieve this by binding the negatively charged groups present on bacterial surfaces and extracellular polysaccharide molecules, facilitating the formation of more cohesive and robust granules. In a separate study, Tsuneda et al. [80] observed that introducing iron (Fe) into the system led to its accumulation in the central part of the nitrifying granules in an AUFB reactor. Furthermore, they found that sludge pre-aggregated with iron could enhance the formation of nitrifying granules. This indicates that iron, like calcium, may play a crucial role in the development and structural integrity of aerobic granules, particularly in systems focused on nitrification. Magnesium carbonate played a similar role in strengthening the formation of aerobic granules [81]. Such insights are valuable for optimizing the process of aerobic granulation in wastewater treatment systems.

8. Application of Aerobic Granulation Technology

Aerobic granulation technology is a versatile and efficient solution for wastewater treatment, offering benefits like a high treatment efficiency, space savings, energy-efficiency, and environmental sustainability. Its application spans across municipal and various industrial sectors, addressing both conventional and challenging wastewater treatment needs [23,40,61,69,82–89].

In treating high-strength organic wastewater, biological systems need to maintain a high biomass concentration and a high rate of microbial degradation. Aerobic granulation in SBRs has proven effective in achieving this, with biomass concentrations between 6.0 to 12.0 g L^{-1} being reported due to the compact and dense structure of the granules. This was highlighted in the research by Tay et al. [23,82].

The practicality of using aerobic granulation technology for treating high-strength organic wastewater was demonstrated by Moy et al. [69]. They found that aerobic granules could withstand a maximum organic loading rate of 15.0 kg COD m^{-3} d^{-1} using glucose as a substrate, while achieving more than 92% COD removal. This high efficiency can be attributed to the granules' compact structure, which also enhances their ability to degrade toxic compounds.

For example, studies by Jiang et al. [61,83] showed that aerobic granules are particularly effective in degrading toxic substances like phenol. In one instance, an aerobic granular sludge reactor successfully maintained a steady effluent phenol concentration below 0.2 mg L^{-1}, despite the influent having a phenol level of 500 mg L^{-1}. This high resilience of the granules to phenol can be attributed to the fact that a significant portion of the biomass inside them remains shielded from the elevated concentrations encountered in the wastewater.

Aerobic granules are gaining recognition for their ability to eliminate not just phenol, but also other challenging and toxic organic substances from concentrated industrial wastewater. Additionally, recent research has shown their ability to efficiently reduce phosphate and ammonia levels [40,84,85]. Their adaptability and effectiveness position aerobic granules as a viable option for sophisticated wastewater treatment endeavors.

9. The Engineering of Aerobic Granular Sludge Technology

Following the first pilot-scale study of aerobic granular sludge reported in 2003, dozens of pilot-scale studies were completed (Table 3). The influent of the pilot-scale reactors include sewage, industrial wastewater, and their mixture. The volume of the reactors ranged from a minimum of 30 L to a maximum of 6 m^3. The inoculation sludge was mostly obtained from the activated sludge of urban wastewater plants, and the raw water included domestic sewage, industrial wastewater, and agricultural wastewater. In the pilot studies, the COD of the raw water ranged from 100 mg L^{-1} to 2000 mg L^{-1}, and $NH_4{}^+$-N was between 10 mg L^{-1} and 200 mg L^{-1}. The size of the cultivated granular sludge ranged from a minimum of 0.2 mm to a maximum of 3.5 mm.

Table 3. The list of pilot-scale aerobic granulation processes.

Year	Reactor	Working Volume (m^3)	Inoculation Sludge	Influence	Flow Pattern	MLSS (g/L)	Diameter of Granules (mm)	SVI (mL/g)	Reference
2003	SBR	1.5	Sludge from municipal wastewater treatment plant	Sewage	Intermittent flow	9–10	>0.6	60	[90]
2010	SBR	0.03	Sludge from municipal wastewater treatment plant, MLSS: 2 g L^{-1}, SVI: 145 mL g^{-1}	40% sewage + 60% industrial wastewater, COD: 360–1832 mg L^{-1}, $NH_4{}^+$-N: 37.5–108.5 mg L^{-1}	Intermittent flow	20	0.8	30	[91,92]
2010	SBR	0.226	Sludge from municipal wastewater treatment plant, MLSS: 2.6 g L^{-1}, SVI: 120–160 mL g^{-1}	Municipal wastewater, COD: 91.3–157.1 mg L^{-1}, $NH_4{}^+$-N: 39.4–68.2 mg L^{-1}	Intermittent flow	4.0	2.45	45–55	[93]
2010	SBR	6.0	Activated sludge, MLSS: 3.0 g L^{-1}	Municipal wastewater, COD: 200–350 mg L^{-1}, $NH_4{}^+$-N: 15–40 mg L^{-1}	Intermittent flow	8.0	0.33	30	[94]
2010	SBR	1.0	Sludge from municipal wastewater treatment plant, MLSS: 5.0–7.0 g L^{-1}, SVI: 75 mL g^{-1}	Municipal wastewater, COD: 100–400 mg L^{-1}, $NH_4{}^+$-N: 10–40 mg L^{-1}	Intermittent flow	8.0	0.8	40	[95]
2011	SBR	0.032	Sludge from municipal wastewater treatment plant, MLSS: 2.6 g L^{-1}, SVI: 180 mL g^{-1}	40% sewage + 60% industrial wastewater, COD: 250–1800 mg L^{-1}, $NH_4{}^+$-N: 39–93 mg L^{-1}	Intermittent flow	7.0–9.0	1.976	25–85	[96]
2011	SBR	0.1	Sludge from municipal wastewater treatment plant, MLSS: 3.7g L^{-1}, SVI: 190 mL g^{-1}	The synthetic wastewater (acetate)	Intermittent flow	3.5	3.5	-	[97]
2011	SBR	5.95	Sludge from municipal wastewater treatment plant, MLSS of 2.7 g L^{-1}	Sewage with industrial wastewater, COD: 271–1839 mg L^{-1}, $NH_4{}^+$-N: 16.98–214 mg L^{-1}	Intermittent flow	2.236	-	65.02	[98]
2012	SBR	1.47	Sludge from soy protein wastewater treatment plant with SVI of 125.6 mL g^{-1}	Soy protein wastewater anaerobic digest effluent with COD of 800–1800 mg L^{-1}, $NH_4{}^+$-N of 80–160 mg L^{-1}	Intermittent flow	-	0.5–1.0	-	[99]
2012	SBR	0.085	Anaerobic digest sludge, MLSS of 20 g L^{-1}	Sewage with COD of 200–320 mg L^{-1}, TN of 38–55 mg L^{-1}	Intermittent flow	5.9	0.75	20–35	[100]

Table 3. *Cont.*

Year	Reactor	Working Volume (m^3)	Inoculation Sludge	Influence	Flow Pattern	MLSS (g/L)	Diameter of Granules (mm)	SVI (mL/g)	Reference
2012	SBR	0.1	-	The synthetic wastewater (Sodium acetate) with COD of 400 mg L^{-1}, NH_4^+-N of 40 mg L^{-1}	Intermittent flow	12±4	2.4	13±6	[101]
2013	SBR	1.47	Activated sludge with MLSS of 2.8 g L^{-1}, SVI of 105.51 mL g^{-1}	Soy protein wastewater anaerobic digest effluent with COD of 700–2400 mg L^{-1}, NH_4^+-N of 200 mg L^{-1}	Intermittent flow	7.02	1.2–2.0	42.99	[102]
2013	MBR	0.06	Sludge from pharmaceutical wastewater treatment plant	Berberine wastewater with COD of 1717–4393 mg L^{-1}, NH_4^+-N of 91.8–158.7 mg L^{-1}	Continuous flow	7.0	0.1–1.0	90	[103]
2013	SBR	0.1	Sludge from municipal wastewater treatment plant	Swine wastewater	Intermittent flow	11–13	2.0–2.8	-	[104]
2014	SBR	3.5	Sludge from municipal wastewater treatment plant with MLSS of 4.581 g L^{-1}	Municipal wastewater, COD of 100–450 mg L^{-1}, NH_4^+-N of 20–30 mg L^{-1}	Intermittent flow	1.2	1.0	-	[105]
2014	SBR	0.105	Sludge cultivated in lab with MLSS of 3.0 g L^{-1}	The synthetic wastewater (Sodium acetate) with COD of 8000 mg L^{-1}	Intermittent flow	5.0	1.58	80	[106]
2014	SBR	20	Activated sludge with MLSS of 3.8 g/L, SVI of 78 mL g^{-1}	30% sewage + 70% industrial wastewater with COD of 500–1000 mg L^{-1}, NH_4^+-N of 30–80 mg L^{-1}	Intermittent flow	8.55	0.3	38	[107]
2015	SBR	4	Sludge from enhanced biological phosphorus removal treatment	Sewage with Sodium acetate	intermittent flow	12	1.1	-	[108]
2016	SBR	1.5	Activated sludge	Sewage	intermittent flow	9–10	>0.6	60	[109]
2016	MBR	14	Sludge from municipal wastewater treatment plant with SVI of 210 mL g^{-1}	Municipal wastewater, COD of 300 ± 25 mg L^{-1}, TN of 30 ± 5 mg L^{-1}	intermittent flow	7	0.2	30	[110]
2017	SBR	0.098	Sludge from municipal wastewater treatment plant with VSS of 3.2 g/L, SVI of 220.2 mL g^{-1}	Sewage with COD of 150–450 mg L^{-1}, NH_4^+-N of 36–68 mg L^{-1}	intermittent flow	-	0.29	67	[111]
2017	SBR	0.16	Sludge from municipal wastewater treatment plant with MLSS of 6.5 g L^{-1}	Municipal wastewater, COD of 300 mg L^{-1}, NH_4^+-N of 43–52 mg L^{-1}	intermittent flow	12.19	1.269	21.31	[112]
2019	CSTR	0.128	Activated sludge with 4.284 g L^{-1}	Sewage, COD of 200–400 mg L^{-1}, NH_4^+-N 0f 10–35 mg L^{-1}, TN of 30–55 mg L^{-1} and TP of 1–5 mg L^{-1}	Continuous flow	4.1–5.8	3.4	64	[113]
2022	RFBR		Activated sludge	-	Continuous flow	1.3	-	43	[18]
2023	SFD-SBR	Sidestream reactor: 1.4 m^3, mainstream reactor: 14 m^3	Sludge from municipal wastewater treatment plant	The municipal wastewater, TOC of 48–59mg L^{-1}	Continuous flow	3	-	80	[114]
2023	CFR	0.2	Sludge from municipal wastewater treatment plant with MLSS of 2.5 g L^{-1}	The municipal wastewater, COD of 161–1145 mg L^{-1}; TN of 14–103 mg L^{-1} and TP of 2.5–19 mg L^{-1}	Continuous flow	6.1	0.5–1.0	40	[115]

The first full-scale AGS reactor was set up in the Garmerwolde Wastewater Treatment Plant in The Netherlands. At present, more than 80 AGS plants are operated worldwide by Nereda®. The operation condition and results of a few of these full-scale reactors, located in Poland, The Netherlands, and South Africa, were published [109–116]. Full-scale aerobic granulation is a complex process that requires careful design, monitoring, and management to successfully treat wastewater in an efficient and environmentally friendly manner. The technology's scalability and adaptability to various wastewater treatment needs make it an increasingly popular choice in modern wastewater treatment facilities (Table 4).

Table 4. The list of full-scale continuous-flow aerobic granulation processes.

Year	Location	Name	Granulation Strategy	Reactor Type	Wastewater Treatment Plant Capacity (m^3/d)	Inoculation Sludge	Influence	MLSS (g/L)	Diameter of Granules (mm)	SVI (mL/g)	Reference
2008	South Africa	Gansbaai WWTP	SBR (Nereda)	Column	4000	Activated Sludge	Sewage, COD: 1265 mg L^{-1}, $NH_4{}^+$-N: 175 mg L^{-1}, TP: 19 mg L^{-1}	-	-	-	[117]
2010	Zhejiang Province	Yancang WWTP	SBR	Column	50,000	Activated Sludge	70% industrial wastewater + 30%municipal wastewater, COD: 200–700 mg L^{-1}, $NH_4{}^+$-N: 28–40 mg L^{-1}, TP: 2–4 mg L^{-1}	-	0.5	47	[13]
2013	The Netherlands	Garmerwolde	SBR (Nereda)	Column	13,000	Activated Sludge	Sewage, COD: 146–715 mg L^{-1}, $NH_4{}^+$-N: 13.4–56.5 mg L^{-1}, TN: 14–81 mg L^{-1}, TP: 1.9–9.7 mg L^{-1}	8.5	1	35	[117]
2014	Portugal	Frielas WWTP	SBR (Nereda)	Column	70,000	Activated Sludge	Sewage	6–8	-	40	[117]
2022	The James R. Dilorio Water Recla mation Facility	Colorado, USA	Hydrocyclone-based wasting helped improve settling characteristics	Several tanks	60,000	Sludge from municipal wastewater treatment plant	Sewage	2.2	>0.2	83	[118]
2024	WWTP in	Hebei province, China	A novel microaerobic–aerobic configuration with internal separators	Several tanks	25,000	Sludge form Municipal wastewater treatment plant	30% sewage + 70% industrial wastewater, COD: 200–700 mg L^{-1}, $NH_4{}^+$-N: 28–40 mg L^{-1}	20	0.138, granules larger than 200 µm constituting 28.9%	51.4	[119]

10. Conclusions and Challenges in Future

This paper offers an extensive review of the advancements in aerobic granular sludge research, highlighting the evolution of aerobic granulation technology, the mechanisms behind granulation, the properties of aerobic granules, the factors that drive aerobic formation, and their practical uses. Despite the growing worldwide implementation of AGS in wastewater treatment plants (WWTPs), indicating a promising future for AGS in boosting wastewater treatment efficiency, several critical challenges hinder AGS application. The key obstacles and future research opportunities in AGS include the following:

(1) The process of aerobic granule formation remains largely unknown. Various theories have been proposed to explain this phenomenon, yet conclusive experimental support is lacking.

(2) Achieving low substrate concentrations during steady-state operations to enable feast/famine conditions in CAGS is difficult. Research into how CAGS's operational parameters affect the feast/famine ratio is scarce, necessitating future investigations into its impact on granule stability.
(3) Employing a particle-size-based selection pressure could potentially facilitate CAGS granulation. However, research in this area is still in its infancy, and such a selection pressure might compromise CAGS stability in the long term.
(4) While most CAGS setups incorporate a settling tank or clarifier for granule recycling, the recycling process can lead to granule disintegration. This calls for the creation of more sophisticated CAGS reactors with optimized recycling mechanisms.

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Abbreviations

ALR	Airlift reactor
CFR	Continuous-flow reactor
COD	Chemical oxygen demand
EPSs	Extracellular polymeric substances
HRT	Hydraulic retention time
MBR	Membrane bioreactor
OLR	Organic loading rate
RFBR	Reverse flow baffled reactor
SBR	Sequential batch reactor
SVI	Sludge volume index
WWTP	Wastewater treatment plan

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