

# REVIEW OF AEROBIC GRANULAR SLUDGE TECHNOLOGY AND APPLICATION IN WASTEWATER TREATMENT

TRUONG THI BICH HONG

*Faculty of Natural Education, Pham Van Dong University*

*\*Corresponding author: ttbhong@pdu.edu.vn*

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**Abstract.** This paper focuses on introducing an overview of aerobic granular sludge technology, granulation, removal efficiency and application of aerobic granules in wastewater treatment. Aerobic granular sludge has a bacterial layered structure including anaerobic, anoxic and aerobic bacteria in order from inside to outside layer, so it is capable of simultaneous chemical oxygen demand, nitrogen and phosphorus removal. Granular sludge also has capacity to remove persistent organic substances and adsorb heavy metals. As a result, aerobic granular sludge technology can be effectively applied for municipal and industrial wastewater treatment. Many wastewater treatment plants applying aerobic granular sludge technology have been deployed, most commonly in Europe and especially in the Netherlands, which is the first place to apply this technology in practical wastewater treatment with Nereda® trademark. However, aerobic granular sludge is stable on batch reactors and takes months to form stable granular sludge. Therefore, it is necessary to have further researches on the granulation mechanism and affective factors to shorten the time of aerobic granular sludge formation as well as maintain the long-term stability of the granular sludge and develop continuous reactors to maintain the stability of the aerobic granular sludge to meet the requirement of wastewater treatment plants in practice.

**Keywords:** aerobic granular sludge, full-scale application, granulation, nutrient removal, wastewater treatment.

## 1. INTRODUCTION

The population growth, urbanization and industrialization have generated many environmental problems that affect the quality of life of most of the inhabitants in the earth. Along with the increasing climate change, the reserve of clean water is being reduced due to saltwater intrusion, environmental pollution, especially wastewater from daily-life activities and factories. To solve this problem, the treatment of wastewater before being discharged into the environment must be carried out thoroughly and effectively and towards wastewater reuse in the future. Therefore, it is required to apply high technologies in wastewater treatment.

Up to now, biological wastewater treatment technology is still considered an effective technology to treat organic components (chemical oxygen demand - COD) and nutrients (nitrogen, phosphorus) in wastewater. In practice, the conventional activated sludge (CAS) technology is being applied commonly in most wastewater treatment systems today such as aerotank, sequencing batch reactor (SBR), membrane bioreactor (MBR)... However, the CAS technology has limitations such as low COD and nitrogen loading, easy to loading shock, sensitive to toxins, ineffective to treat COD and nitrogen simultaneously, small sludge size causing membrane fouling [1-3].

The development of aerobic granular sludge (AGS) technology overcomes the limitations of CAS technology due to advantages such as high settling velocity, capacity to withstand to high organic loading rate (OLR) and low sensitivity to toxins [4, 5]. AGS has a specific structure consisting of an inner anaerobic sludge layer and an outer aerobic sludge layer, so COD and nitrogen can be treated simultaneously if dissolved oxygen (DO) concentration are controlled at a low level and the sludge particle size is large enough to create an anoxic zone inside the granular sludge [6, 7]. Furthermore, AGS with a dense structure and large particle size will be the solution to reduce membrane fouling in MBR [8, 9].

With the above advantages, AGS technology has been researched and developed, many practical applications have been deployed in the world. However, the studies on AGS technology are still restricted in Vietnam. To have a systematic view of AGS technology and its applications, this review will focus on: (1) overview of the AGS technology, (2) the capacity to treat pollutants in wastewater of AGS, (3) applications, challenges and further investigations of AGS technology. The novelty of this study is to

evaluate operating conditions and modes affecting to the granulation and determine suitable operating parameters. The treatment efficiency of organic substances and other components such as metals, persistent organic matters is also evaluated in this study. This information is very useful for establishing the operating procedure of the AGS process and applying AGS in the treatment of various types of wastewater.

## 2. OVERVIEW OF THE AEROBIC GRANULAR SLUDGE TECHNOLOGY

### 2.1. Introduction about aerobic granular sludge

#### 2.1.1. The structure of aerobic granular sludge

Aerobic granules are aggregation of suspended biomass, including bacterial cells, inert particles, easy-degradable particles, and extracellular polymeric substances (EPS) (Figure 1). Among them, EPS acts as a binder of the components together and under good mixing conditions of the reactor they will be rounded and form into granules. Granular sludge is a special case of biofilm in which microorganisms bind to form granular sludge without a carrier material.

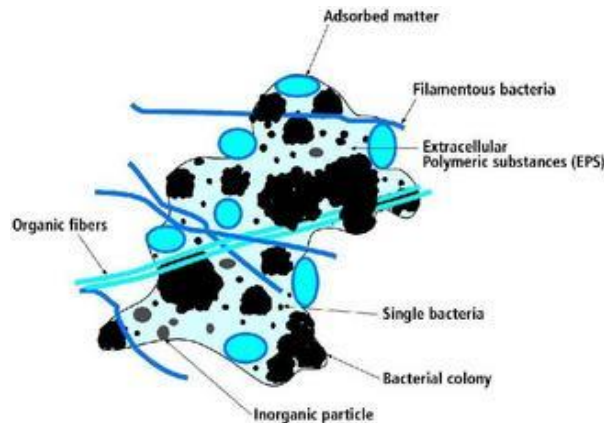


Figure 1: Biomass binds together to form granules [10]

Due to limited oxygen diffusion, AGS has a layered structure with the presence of different bacterial groups. The innermost layer is core, which contains dead cells, inert substances and anaerobic bacteria and creates the sludge center structure; the next layer is a class of anoxic bacteria such as denitrifying bacteria and phosphate accumulating organism (PAO) and the outermost layer consists of aerobic bacteria such as ammonia-oxidizing bacteria and protozoa mainly ciliates and rotifers. The thickness of the anaerobic and anoxic layers decreases with the size of the granule. The granular sludge structure has many pores, so that the substrate can diffuse into the granular sludge, and the toxic substances generated during metabolism can also be easily separated from the granular sludge. Bacterial structure of AGS is shown in Figure 2.

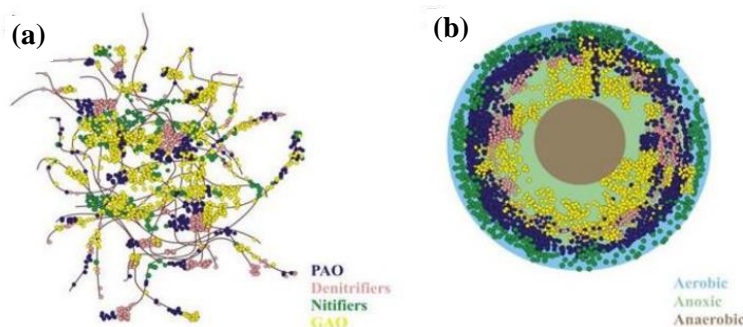


Figure 2: Bacterial structure of aerobic granular sludge: (a) Floc; (b) Granule [11].

#### 2.1.2. The characteristic of aerobic granular sludge

The granular sludge has the characteristic of aerobic sludge in the outer layer and anoxic sludge in the inner layer, so it can remove COD and nitrogen simultaneously if DO concentration is limited to a certain level and the sludge size is large enough to create an anoxic zone inside the granular sludge. The treatment capacity and stability of AGS depend on the physical and biological properties of the sludge. The most

important physical properties of granular sludge are particle size, settling capacity and mechanical strength of granular sludge. The biological properties are determined mainly by the composition and number of microbial species present in the granular sludge. AGS has distinct properties and advantages over CAS and is compared in Table 1.

Table 1: Comparison of properties of aerobic granular sludge and conventional activated sludge.

Aerobic granular sludge [12]	Conventional activated sludge [13]
- The surface of granule is even and smooth	- The surface of floc is uneven and unsmooth
- Granule is heavy and compact	- Floc is light and loose
- Low settling time (2-3 minutes)	- High settling time (2-3 h)
- High biomass concentration (>10 gVSS/L)	- Low biomass concentration (<5 gVSS/L)
- High organic loading rate (5-8 kgCOD/m <sup>3</sup> .day)	- Low organic loading rate (<2.5 kgCOD/m <sup>3</sup> .day)
- Less loading shock	- Easy to loading shock

## 2.2. Aerobic granulation process

### 2.2.1. Mechanism of aerobic granular sludge formation

The fact that formation of granular sludge is a natural process. The aerobic granulation process is essentially the process of self-adhesion of bacteria together. This phenomenon is common in all biotechnological wastewater treatment systems that meet basic operating conditions such as high hydraulic shear force (corresponding to the surface air velocity 1-3 cm/s [14]), short settling time (corresponding to the settling velocity of sludge particles higher than 10 m/h [15]), appropriate substrate loads (5-8 kgCOD/m<sup>3</sup>.day [16]). The mechanism of AGS formation in a sequencing batch airlift reactor (SBAR) is similar to that in a sequencing batch reactor (SBR) or a suspended biofilm reactor using a continuous airlift (BAS) [15]. Different hypotheses have been proposed to explain the mechanism of aerobic granulation. However, the most common hypothesis proposed was based on experiments with above specific conditions as illustrated in Figure 3 and gets agreement of many authors [12, 16-18].

According to Figure 3, under good disturbance of the gas flow, the filamentous bacteria bind together and form flocs with high settling velocity, which should be kept in the reactor. Initially, the bacteria can't adhere to each other, so they have a low settling velocity and are washed out of the reactor. Therefore, during the acclimatization phase the flocs are mainly composed of filamentous bacteria. Due to the shearing force, the filamentous bacteria gradually separate from the surface of the floc, making the floc become stronger and larger up to a size of about 5-6 mm. These flocs will form a surface that attracts other bacteria to attach and grow into clusters of bacteria. However, due to the limitation of oxygen diffusion inside the flocs, these flocs are gradually broken up and washed out in the effluent, and the bacteria are now clustered, so the settling velocity is large enough to retain in the reactor. These bacterial clusters will develop into granular sludge and gradually dominate in the reactor.

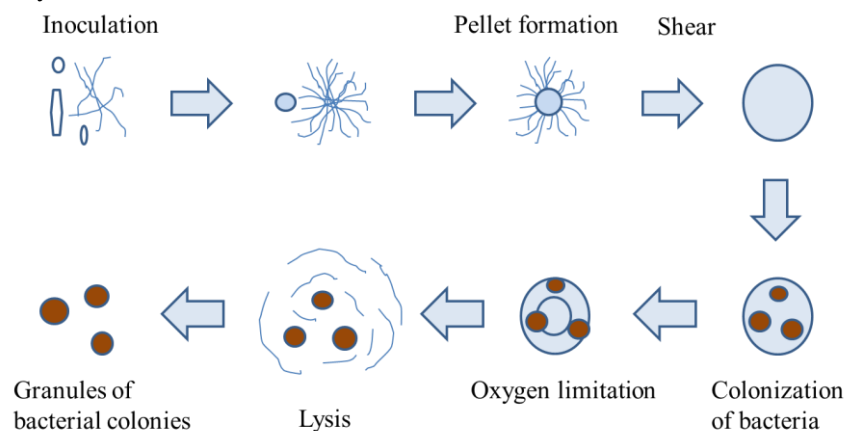


Figure 3: Mechanism of aerobic granulation in SBR [17].

The result in Figure 4 shows that the AGS formation process consists of 3 stages: acclimatization phase, granulation stage and maturation stage [19]. Acclimatization phase is from the start up of the reactor until the formation of granular germ. The granular germ is mainly composed of adherent filamentous bacteria. Granulation stage is from the formation of granular germ to the formation of granules including many coexisted various types of bacteria and the biomass concentration in the reactor is stable. At maturation stage, the granular sludge grows rapidly in size and density after only a few weeks, while the floc will be washed out of the reactor. After about 7 weeks, the granular sludge forms almost completely and there is very little suspended biomass in the reactor. After the growth point, the granular sludge will stabilize and balance, maintaining the particle size in the range of 2-5 mm. Many researches with above suitable operational conditions also show the same aerobic granules formation as listed in Table 2.

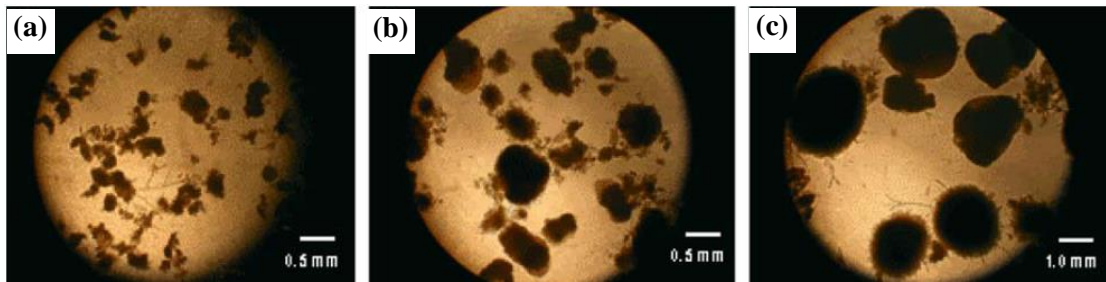


Figure 4: The formation and development of aerobic granular sludge: (a) granular germ; (b) granular formation; (c) mature granules with clear borders [19].

### 2.2.2. Factors affecting the aerobic granulation process

Granular sludge can be formed by many different groups of microorganisms. It is clear that the granular sludge formation is not restricted by microbial groups but is related to the operating conditions in the reactor [17]. The AGS formation is mainly related to the operating conditions and modes in the reactor such as settling time, hydraulic retention time (HRT), OLR, gas flow rate and DO concentration, shape of the reactor and wastewater properties. In which, the operating parameters and modes as well as wastewater properties directly affect to the granulation, while the reactor shape indirectly affects the granulation through hydraulic shear force. The hydraulic shear force is determined by the gas flow rate and the reactor shape and is considered to be the main factor that affect to the aerobic granulation. Factors affecting the aerobic granulation is shown in Figure 5.

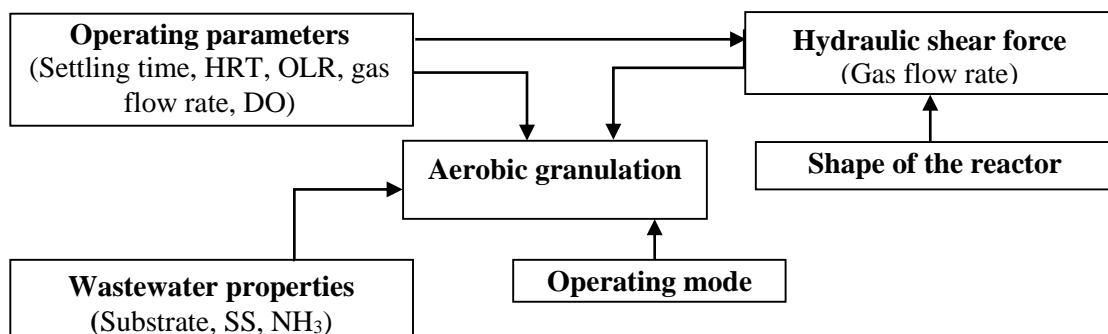


Figure 5: Factors affecting the aerobic granulation process.

The operating parameters and conditions of AGS process is listed in Table 2. The data showed that settling time is a very important factor in granular sludge formation because it determines the amount of sludge accumulated in the reactor. The selection of sludge particles to retain in the reactor is based on the different settling rates between the granular sludge and the floc. If the settling time is long, only floc sludge will

develop in reactor, on the contrary, if the settling time is too short, it will wash away the particles which also have good settling ability. In AGS formation, the settling time should be chosen so that the settling velocity of sludge particles higher than 10 m/h [15].

HRT is also a main factor that directly affect to the granulation. The data in Table 2 shows that the SBR is operated for only a few hours (3-6 hours) corresponding to HRT of about 6-12 hours. The substrate will be rapidly consumed during the first 20-60 minutes of the aeration cycle. Therefore, increasing OLR and decreasing HRT will increase the growth rate of bacteria and generate more EPS, which plays the role of sticking bacterial cells together, increasing the growth rate of granule sludge and making granular sludge to be denser [16]. Many studies concluded that small HRT is beneficial for granulation [17, 20].

Studies showed that AGS is synthesized at high OLR and the floc exists mainly at low OLR. The floc has a loose and irregular structure with a small size, therefore it has a low substrate diffusion limit leading to have favorable conditions to compete and grow at a lower load than the granular sludge. Furthermore, high OLR promotes the production of EPS, which binds bacterial cells to enhance granular sludge formation. At low OLR, granular sludge is difficult to form or if it is, it takes a long time [21, 22] and the granule is small [23]. However, a too high OLR will increase the growth of filamentous bacteria and reduce the settling capacity of the granular sludge. Therefore, to maintain the stability of the granule sludge, it is necessary to control OLR in the appropriate range of about 5-8 kgCOD/m<sup>3</sup>.day [16]. The effect of various OLRs to granular size is shown in Figure 6.

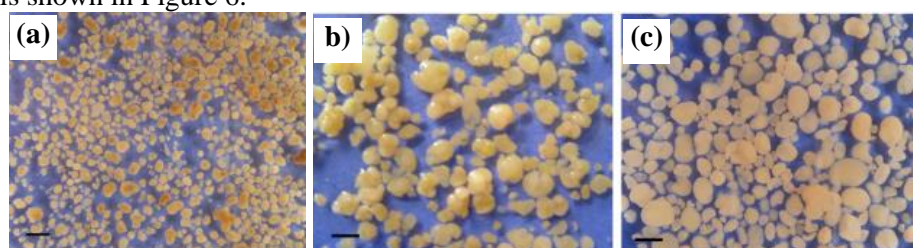


Figure 6: Mature granules at various organic loading rates: (a) 1.5 kgCOD/m<sup>3</sup>.day; (b) 3 kgCOD/m<sup>3</sup>.day; (c) 4.5 kgCOD/m<sup>3</sup>.day [24].

The gas flow rate affects the granulation by determining the DO concentration and hydraulic shear force in the reactor. The shear force is generated by the friction between the liquid, gas and solid particles and is represented by an increase in the surface gas velocity in the reactor [14]. High shear force will increase the hydrophobicity of the granules, which is one of the factors determining the uniform shape and density of the granules. Moreover, when the shear force is high, it will increase the metabolism of microorganisms and secrete more EPS, increasing the ability to stabilize the structure of the granular sludge [14]. However, when the gas flow is too high, it will break the granular sludge structure. To ensure favorable conditions for aerobic granulation, the gas flow rate is selected in the range of 2-6 L/min (corresponding to the surface air velocity 1-3 cm/s) [14, 25]. AGS is unstable at low DO concentrations (40% saturation) because when DO is low, filamentous bacteria will grow and cause the granular sludge to decompose [26]. Therefore, DO in the reactor must be guaranteed to be higher than 2 mg/L.

The operating mode directly affects the supply and consumption of substrates by microorganisms in the reactor. Batch-operated reactors (SBR) with cycles including filling, aeration, settling and draining stages create alternate feast and famine conditions in the reactor thereby promoting production and consumption of EPS compounds to bind bacterial cells to form granules. Moreover, the high substrate concentration in the filling stage can diffuse deep into the granular sludge (500 μm), thereby maintaining the growth and development of bacteria inside the sludge [15, 27].

The shape of the reactor indirectly affects the aerobic granulation through hydraulic shear force. Studies show that column reactors with a high H/D ratio give higher hydraulic shear forces with the same gas flow and thus promote granulation [14]. Furthermore, a reactor with a high H/D ratio will result in higher settling velocity at the same discharge and settling times. An H/D ratio of about 10-20 is often used to provide the minimum settling velocity for granulation [15, 17]. In practice, many continuously operating reactor are used, so studies have tried to create and maintain the stability of granular sludge on continuous operating models to meet practical requirements [28-30]. However, the results are also only at the research and development phase.

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Property of wastewater also affects to the aerobic granulation. AGS can grow on many different substrates with high organic matter content such as synthetic wastewater (acetate, glucose, peptone, sucrose, ...) [16, 25, 31], domestic and industrial wastewater [32-34]. However, the structure and microbial composition in granule sludge cultured with different substrates will be different.

Indeed, there are many factors that influence to AGS formation. To deeper understand the mechanism of AGS formation and stabilization, in-depth studies on the influence of factors on the granulation need to be further studied.

Table 1: Overview of operational parameters and treatment efficiency of aerobic granular sludge reactors.

Reactor	Wastewater	Operating parameters					Property of aerobic granules	Removal efficiency (%)					Reference
		Settling time, min	Gas flow rate, L/min (gas velocity, cm/s)	OLR (NLR), kg/m <sup>3</sup> .day	Cycle time (HRT), h	Exchange ratio, %		COD	NH <sub>4</sub> <sup>+</sup>	T-N	T-P (PO <sub>4</sub> <sup>3-</sup> )	Other substances	
<b>SBR</b> V=2.2 L H/D=24	Ethanol	2	(4.1)	5	3 (6.75)	50	d <sub>ib</sub> = 3.3 mm						[17]
<b>SBAR</b> V = 3 L H/D = 11.25	Natri acetace	3	4.4 (2.22)	2.5	3 (5.6)	50	d <sub>ib</sub> = 2.5 mm						[15]
<b>SBR</b> V = 2.4 L H/D = 24	Glucose Acetace	20→1	3 (2.5)	6	4	50	Glucose: d <sub>ib</sub> = 2.5 mm, Acetate: d <sub>ib</sub> =1.1 mm, dense	97- 98					[31]
<b>SBAR</b> V = 3.5 L H/D = 11.25	Sucrose	5	2	1.76	3	50	d <sub>ib</sub> = 1.5 mm						[19]
<b>SBAR</b> V = 3 L H/D = 15.6	Sodium Acetace	3	4	1.2-1.6	3 (5.6)	50	d <sub>ib</sub> =1.1 mm			44-75	(95-97)		[35]
<b>SBR</b> V = 8.6 L	Brewery wastewater	4	(1.77)	3.5 (0.24)	6	50	D = 2-5 mm	88.7	88.9				[32]
<b>SBR</b> V = 2 L H/D = 24	Acetace Domestic wastewater	30→10	3.5 (3)	0.6-1.2	4→2	50	d <sub>ib</sub> = 0.72 mm SVI = 65 mL/g MLVSS = 2 g/l	> 90	90				[1]
<b>SBR</b> V = 5 L	4-chloroaniline (4CIA)		(2.4)	4CIA: 20 g/m <sup>3</sup> .day	(17.2)		d <sub>ib</sub> = 1.67 mm	93		70		99.9	[36]
<b>SBR</b> V = 32 L	Domestic, industrial wastewater	10→2	1.6			50-75	d <sub>ib</sub> = 2.5 mm (day 48)	80	90				[37]
<b>SBR</b> V=100 L	Swine slurry wastewater	10→4		1.4-6.3 (0.5-2.5)	3(6)	50	d = 1-3 mm	61-73	56-77				[38]
<b>SBR</b> V = 2 L H/D= 20	Sodium Acetate	19	(1.25)	2.69	6 (10)	50	d <sub>ib</sub> = 600 μm (day 3)						[16]
<b>SBR</b> V = 3.3 L	Tapioca processing wastewater	3	5 (2.5)	2.5-10	3 (5.67)	50	d <sub>ib</sub> = 2.5 mm	90- 93	86-92	66.1			[12]
<b>SBR</b> V = 1 L	Azo dye wastewater (Mordant Orange 1)	1	3	Mordant Orange 1: 0.1g/L		50	d = 0.3-2.4 mm	> 90				61-88	[39]



<b>SBR</b> V=120 L H/D = 6	Septic tank sewage	2	(2.4)	8	2 (4)	60	d = 2-3 mm, granules stabilize for long time	> 90		>70	>90		[27]
<b>SBR</b> H/D = 15 H = 50cm	Synthetic swine wastewater	1	1.4	COD= 3000 mg/L	(4.8-16)		= 2-3 mm	> 95	88-93		70-89		[40]
<b>SBR</b>	Domestic wastewater			0.9±0.3			d > 0.2 mm	80	83		(55)		[23]
<b>SBAR</b> V=5.4L		5			6	50					94		[41]
<b>SBR</b> H/D=10			3.5			50		96.1	99.6		98		[42]

Note: OLR: organic loading rate, kgCOD/m<sup>3</sup>.day; NLR: nitrogen loading rate, kgN/m<sup>3</sup>.day, T-N: total nitrogen, T-P: total phosphorous.



### 3. CAPACITY OF POLLUTANT REMOVAL AND MEMBRANE FOULING REDUCTION OF AEROBIC GRANULAR SLUDGE

#### 3.1. Removal of organic compounds and nutrients

Due to the diffusion limitation, during the substrate-rich phase, oxygen can only diffuse into the granular sludge to a depth of 150-350  $\mu\text{m}$  [6, 43]. Therefore, the large granular sludge (1-5 mm) with a special structure consisting of an outer aerobic zone and an inner anoxic zone is capable of simultaneous organic matter, nitrogen and phosphorus removal. The outer layer of the granular sludge concentrates mainly aerobic bacteria including heterotrophic bacteria that decompose organic compounds and nitrifying bacteria that oxidize ammonium to  $\text{NO}_3^-$ . Inside the granular sludge concentrates mainly anoxic and anaerobic bacteria such as denitrifying bacteria and PAO bacteria. The denitrifying bacteria use electron acceptors that are organic substances in the input wastewater or organic substances generated during intracellular decomposition or added from the outside to perform the denitrification. Phosphorus in the wastewater is absorbed into the biomass cell of the PAO bacteria, which is then removed through the residual sludge discharge.

The data in Table 2 show the organic compound and nutrient removal capacity of AGS process. The AGS can remove COD at high organic loads of 2.5-10  $\text{kg COD/m}^3\cdot\text{day}$  [12]. The COD removal efficiency reaches more than 90% in the reactor with high sludge concentration (5-20  $\text{gVSS/L}$ ). The granular sludge system can maintain nitrogen loads in the range of 0.1-1.5  $\text{kgN/m}^3\cdot\text{day}$ . The ammonia removal efficiency of aerobic granular sludge reactor is also high about 90%. Nitrogen can be removed by the simultaneous nitrification and denitrification processes in the reactor with the efficiency of 60-70%. Aerobic granular sludge was applied for swine slurry wastewater treatment in pilot scale, however high fraction of non-biodegradable organic matter in the feeding up to 80% decreased COD and ammonia removal with the efficiency of 61–73% and 56–77%, respectively [38]. To improve the nitrogen removal efficiency, there are solutions such as alternating anoxic/aerobic phases [44]; operation with low DO concentration period [15]; supplementing an external carbon source [44].

The total phosphorus removal efficiency of the AGS system is about 55-70%. Total phosphorus removal efficiency can be enhanced up to 90% when advanced phosphorus treatment is applied by alternating anaerobic/aerobic conditions [27, 41, 42]. Anaerobic conditions can be created by static filling or gentle mixing. The phosphorus released during the anaerobic filling phase is then rapidly consumed during the aeration phase and removed through residual sludge discharge [45]. Thus, aerobic granular sludge can remove COD, nitrogen and phosphorus simultaneously, even aerobic condition exists in the reactor. Removal efficiency depends on the structure and size of the granules, DO concentration, external carbon source and bacteria in granular sludge.

#### 3.2. Removal of persistent organic compounds

Aerobic granular sludge not only removes biodegradable organic compounds such as glucose, acetate, peptone, but also degrades persistent organic substances such as phenols [46] and p-nitrophenol (PNP) [47]. The AGS was capable of treating wastewater with a phenol load up to 2.4  $\text{kg/m}^3\cdot\text{day}$  [46]. The granular sludge was not significantly affected by phenol toxicity at an average load of 0.6-1.2  $\text{kg/m}^3\cdot\text{day}$ , except for a slight decrease in phenol degradation in the early stages at load 2.4  $\text{kg/m}^3\cdot\text{day}$ . However, the granules could then acclimate and stabilize quickly after a week [46]. PNP degradation capacity was 19.3  $\text{mg/gVSS}\cdot\text{h}$  at PNP concentration up to 40.1  $\text{mg/L}$  [47]. Granular sludge could also effectively remove pyridine at concentrations of 200-2500  $\text{mg/L}$ . The pyridine degradation rates were 73.0 and 66.8  $\text{mg/gVSS}\cdot\text{h}$  at pyridine concentrations of 250 and 500  $\text{mg/L}$ , respectively [48]. The aerobic granules tended to be mature in reactors operated with 4-CIA loading rate of around 800  $\text{g/m}^3\cdot\text{day}$ , and the removal efficiency of 4-CIA was 99.9% [36]. Aerobic granules removed 61-88% Mordant Orange 1 in azo dye wastewater [39]. Although there are many studies of persistent substance removal but these studies are mainly carried out at the lab-scale so further studies need to be conducted on a larger scale.

#### 3.3. Removal of heavy metals

The biosorption of heavy metals by AGS takes place both on the surface and in the core of the granules and influences to functional groups such as alcohol, ether and carboxyl groups [49]. Investigation of  $\text{Cd}^{2+}$  heavy

metal adsorption by AGS was first studied by Liu, Y. and the results showed that the granules can remove  $\text{Cd}^{2+}$  with the range of 43-566 mg  $\text{Cd}^{2+}/\text{g}$  [50]. The maximal  $\text{Ni}^{2+}$  adsorption capacity of the granules was 35 mg  $\text{Ni}^{2+}/\text{g}$  at pH 6 [51]. The maximum adsorption ability of  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$  by aerobic granules was 246.1 mg  $\text{Cu}^{2+}/\text{g}$  and 180 mg  $\text{Zn}^{2+}/\text{g}$ , respectively [52]. The specific  $\text{Cd}^{2+}$ ,  $\text{Cu}^{2+}$ , and  $\text{Ni}^{2+}$  biosorption capacities of aerobic granular sludge were 1.607, 1.403 and 0.764 meq metal/g, respectively [53]. The AGS also had the ability to adsorb Co and Zn with efficiencies of 55.25 mg Co/g at pH 7 and 62.50 mg Zn/g at pH 5, respectively [54]. Optimization of  $\text{Pb}^{2+}$  removal was done by Liu, M. with the maximum biosorption capacity of 79.58 mg  $\text{Pb}/\text{g}$  at 25°C [55]. Although heavy metal adsorption studies have been conducted for various metals, the studies have mainly been carried out on synthetic wastewater. In order to apply AGS for heavy metal removal in practice, further researches on real wastewater needs to be done.

### 3.4. Capacity of membrane fouling reduction

Using aerobic granular sludge instead of activated sludge can reduce membrane fouling in MBR. This experiment was reported firstly by Li, Xiufen in 2005 [56] and attracted the interest of researchers. Most of studies showed that aerobic granules with large size and compact structure have better filterability than floc. During filtration 180 days, Tu, Xiang [8] showed the better filterability of aerobic granular MBR (AGMBR) when at the TMP lower than 8 kPa the specific flux still reached more than 1.3 L/m<sup>2</sup>.h.kPa. At high flux of 20 L/m<sup>2</sup>.h, Wang, Yaqin [9] observed the filterability increased according to the diameter of granules as it took 10, 14, 19, 61 days for the critical TMP of 50 kPa was exceeded for block sludge, floc, small granules and large granules, respectively. Aerobic granules were stable with the large size 2.1-2.3 mm resulted in the decrease in fouling in AGMBR [57]. When the amount of granular sludge in the reactor increased, the permeate flux gradually elevated and fouling resistance decreased [58]. The same results were given by other authors [58-60]. Currently, the studies of AGMBR technology are mostly carried out in synthetic wastewater such as glucose [59, 61], acetate [62-64], methanol [65]... There are some studies in real wastewater such as paper mill wastewater [66], Ilsan wastewater [67], municipal wastewater [68]. Studies on pilot scale have also been carried out such as refractory wastewater treatment in a membrane manufacturer and a satisfactory nutrient removal capacity was achieved in this system [69]. The integration of AGS with a gravity-driven membrane system at a pilot scale demonstrated superior nutrient removal [70]. In practice, an SBR could be easily coupled with an ultrafiltration installation in order to be transformed into a side-stream MBR and the result showed that the better the granule's structure, the less the fouling rate and the permeability loss [71]. Further research about long-term filtration operations at full scale, as well as the stability of AGS is recommended.

## 4. APPLICATION, CHALLENGES AND FURTHER INVESTIGATIONS OF AEROBIC GRANULAR SLUDGE TECHNOLOGY

In the early 1990s, AGS technology was born with the first research conducted by Mishima and Nakamura in 1991 on an aerobic upflow sludge blanket (AUSB) [72]. Granulation was also carried out in other reactors such as BAS [73], SBR [17, 20], and SBAR [25, 74]. In recent years, many researchers have been interested in application AGS in wastewater treatment. Due to the change in real wastewater composition and property, the initial AGS studies were mainly conducted on synthetic wastewater from different carbon sources such as glucose, sucrose, acetate, ethanol... [75-79]. To evaluate the applicability of aerobic granular sludge, many studies have been carried out on real wastewater such as domestic wastewater [22, 80], municipal wastewater [81], industrial wastewater [32, 82, 83], landfill leachate [84] and wastewater contains harmful substances such as phenol, tert-butyl alcohol, p-nitro-phenol [47, 85, 86], and heavy metals [52, 54, 55].

Application of AGS for real wastewater treatment on larger-scale reactors has also been deployed such as the pilot scale reactors for domestic and industrial wastewater treatment [37, 87]. The study showed that AGS formed quickly after about 2 weeks and stabilized after 2 months of operation with COD and  $\text{NH}_4^+$  removal efficiency reaching 80% and 90%, respectively [37]. The long-term investigation of a pilot-scale continuous flow reactor using AGS to treat municipal wastewater achieved the best-performance with  $\text{NH}_4^+$  removal of 99% [88]. Pilot research with Nereda® AGS technology was also performed at five WWTPs in the Netherlands focused on granulation and nutrient removal [89] and other studies such as swine slurry wastewater treatment [38] and rapid granular cultivation in a pilot scale SBR [90]. Aerobic granular sludge

has also been used to treat municipal and industrial wastewater on a practical scale. The world's first full scale AGS plant used Nereda® technology and was started up in 2011 at Epe, The Netherlands [89]. The granular sludge with good settling capacity (SVI 47.1 mL/g) and particle size 0.5 mm was formed in aerobic granular SBR at Yancang wastewater treatment plant, China for treating domestic wastewater (30%) and industrial wastewater (70%) [91]. Many applications of AGS technology have been continued and high efficiency of organic matter, nitrogen, and phosphorus removal has been attained [92, 93].

Really, over more than 20 years of research and development, the AGS technology has been widely applied in wastewater treatment and commercialized by RoyalHaskoningDHV with the trademark of Nereda®. This technology is widely applied in Europe, especially in the Netherlands, which is the first place to install a WWTP using AGS technology of Nereda. Many AGS-based plants are installed for treatment of municipal wastewater, industrial wastewater, or a combination of municipal and industrial wastewater treatment [94]. Statistical data show that the number of plants applying AGS technology has increased over time, with 13 plants built in the period 2020-2021 and 11 plants currently in the design phase [94]. With such a popular development, AGS technology has the ability to replace CAS technology to meet increasingly strict discharging standards and towards wastewater reuse. However, the application of AGS technology in practice still has the following challenges:

- Due to the intracellular decomposition process, there is always a small amount of floc with low settling velocity in the AGS reactor. Therefore, further studies are necessary to increase the proportion of granular sludge in the reactor.
- The AGS has many advantages over CAS. However, the time to form stable AGS and the time to restart the AGS reactor are longer than that of activated sludge. Therefore, the mechanism of AGS formation and the effects of operating factors on the stability of granular sludge still need to be further studied in order to control the process well.
- The AGS has the ability to remove persistent organic matter as well as to adsorb heavy metals. However, the studies are mainly conducted on synthetic wastewater and on lab and pilot scales. Therefore, further studies on real wastewater and on a larger scale are necessary to develop practical applicability.
- At present, the AGS granulation is mainly deployed on the SBR batch reactor. However, the reactors in practice mainly operate in continuous mode. Therefore, it is necessary to develop continuous mode AGS reactors to meet practical requirement.

## 5. CONCLUSIONS

Main factors affecting to the AGS process including of settling time, HRT, OLR, gas flow rate, DO concentration, shape of the reactor and wastewater properties. Suitable operating conditions help to produce large and dense aerobic granules. The mature granular sludge can remove COD and nitrogen, phosphorus simultaneously with the efficiency of more than 90%, 60-70%, 50-70%, respectively. With the advantages of aerobic granular sludge such as good settling capacity, high biomass density, capacity of simultaneous organic matter and nutrients removal, the AGS technology has the ability to increase capacity of WWTPs, meet increasingly stringent discharged standards and towards the reuse of wastewater. Aerobic granular sludge technology has many advantages compared to conventional activated sludge technology, so it has been widely applied in the world. Many AGS-based WWTPs developed worldwide for municipal and industrial wastewater treatment. However, there are still challenges that limit the application development of the technology therefore further research is needed to expand its application in practice. More investigation should focus on the granulation mechanism and affective factors; capacity to remove persistent organic and adsorb heavy metal in real wastewater for larger scales; developing continuous reactors suitable for formation and stabilization of AGS.

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## CÔNG NGHỆ BÙN HẠT HIẾU KHÍ VÀ ỨNG DỤNG TRONG XỬ LÝ NƯỚC THẢI

TRƯƠNG THỊ BÍCH HỒNG\*

*Khoa Sư phạm Tự nhiên, Trường Đại học Phạm Văn Đồng*

*\*Tác giả liên hệ: ttbhong@pdu.edu.vn*

**Tóm tắt.** Bài báo tập trung giới thiệu tổng quan về công nghệ bùn hạt hiếu khí, quá trình tạo bùn hạt, hiệu quả xử lý và ứng dụng của bùn hạt hiếu khí trong xử lý nước thải. Bùn hạt có cấu trúc vi khuẩn phân lớp bao gồm lớp vi khuẩn kỵ khí, thiếu khí và hiếu khí theo trình tự từ trong ra ngoài vì thế có khả năng xử lý đồng thời COD, nitơ và photpho. Bùn hạt hiếu khí cũng có khả năng xử lý các chất hữu cơ khó phân hủy và kim loại nặng. Nhờ đó, công nghệ bùn hạt hiếu khí có thể ứng dụng hiệu quả để xử lý nước thải đô thị và nước thải công nghiệp. Nhiều nhà máy xử lý nước thải áp dụng công nghệ bùn hạt hiếu khí được triển khai, chủ yếu ở Châu Âu, đặc biệt là ở Hà Lan là nơi đầu tiên ứng dụng công nghệ này trong xử lý nước thải thực tế với thương hiệu Nereda®. Tuy nhiên, bùn hạt hiếu khí ổn định trên bề phản ứng theo mẻ và phải mất vài tháng để hình thành bùn hạt ổn định. Do đó, cần có thêm các nghiên cứu sâu hơn về cơ chế tạo bùn hạt hiếu khí và các yếu tố ảnh hưởng để rút ngắn thời gian hình thành bùn hạt cũng như duy trì sự ổn định của bùn hạt trong thời gian dài, đồng thời cần phát triển các bề phản ứng liên tục có khả năng duy trì bùn hạt ổn định để đáp ứng yêu cầu của các nhà máy xử lý nước thải hiện có trong thực tế.

**Từ khóa:** bùn hạt hiếu khí, ứng dụng thực tế, quá trình tạo bùn hạt, xử lý dinh dưỡng, xử lý nước thải.

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