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Nutrient removal and energy consumption of aerobic granule system treating low-strength wastewater at low dissolved oxygen conditions

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ABSTRACT

This study evaluated the properties and pollutant removal ability of sludge from low-strength synthetic wastewater during the formation of aerobic granules at low dissolved oxygen (DO) (1.0 ± 0.2 mg L^{-1}). After 141 d operations, the aerobic granules were successfully cultivated. The removal efficiencies of chemical oxygen demand, NH_4^+, total nitrogen and total phosphorus were 91.0%, 98.6%, 77.6% and 92.5%, respectively. The morphology of the mature granules was almost spherical, with an average granule size of 436.5 ± 12 μm. Filamentous bacteria on the surface were embedded in extracellular polymeric substances. The protein amount (190.4 mg g^{-1} VSS) was greater than that of polysaccharide (30.0 mg g^{-1} VSS). Aerobic granule at low DO (1.0 ± 0.2 mg L^{-1}) consumed a 27.9% less aeration rate compared with conventional biological treatment methods. The good removal performance of aerobic granule at low DO could be suitably applied in wastewater treatment with low energy consumption.

Keywords: Aerobic granules; Low dissolved oxygen; Nutrients removal performance; Aerobic granulation; Sequencing batch reactor (SBR); Energy consumption

1. Introduction

Compared with activated sludge, aerobic granule has multiple advantages such as good settling properties, high biological activity and strong capacity of withstanding shock loading [1–3]. Hence, aerobic granular technology is considered a sustainable and cost-effective method to deal with a variety of wastewater [4–6]. The characteristics and stability of aerobic granules are directly decided by their growth environment, including dissolved oxygen (DO) [7], temperature [8], organic loading [9], etc. The current studies are mainly focused on the optimization of oxygen supplied in aerobic granule processes [10,11]. Yuan and Gao [10] reported that DO of 2.5 mg L^{-1} in aerobic granular sludge reactor made good nitrogen and organic carbon removal. Wang et al. showed that the chemical oxygen demand (COD) removal at DO among 1.0–4.5 mg L^{-1} ranged from 88.9% to 92.2% and increasing the DO could promote the specific ammonium oxidation rate [12]. However, nitrogen removal was favored by low oxygen concentrations [13]. Moreover, the higher DO, the better activity of glycogen accumulating organisms (GAOs), which decreased the *P*-removal [14]. At low DO, polyphosphate accumulating organisms (PAOs) can outcompete GAOs [15]. However, a study on the aerobic granule processes on nutrient removal under low DO condition are still sparse.

The aeration system is an important part of wastewater treatment, which transfers oxygen into a liquid solution using an aeration device [16,17]. The blower in the aeration system is a kind of high-power construction equipment, and its energy consumption is huge. Henriques et al. presented the aeration system accounted for 53% of the overall electric

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energy consumption and more than 12% of pumping operations [18]. Therefore, reducing aeration could lower the energy consumption of wastewater treatment systems. So any effort to reduce DO could realize significant benefits. Fan et al. [19] presented several methods of wastewater treatment, which reduced energy consumption focusing on low DO and low mixed liquor, biogas production. Besides its important role in the granulation process, DO also determines the aeration rate of an aeration system. Under the condition that sufficient nutrient removal was assured, decreasing DO could save energy consumption. The objective of this work was to investigate aerobic granulation at low DO conditions, and offer useful information about the properties and pollutant removal ability of aerobic granule for actual engineering. Meanwhile, the energy consumption calculation of the aeration system was also studied.

2. Materials and methods

2.1. Seed sludge and influent

Seed sludge was from an aerobic tank at Taiping WWTP in Harbin, Heilongjiang Province, China. The concentrations of mixed-liquor suspended solids (MLSS) concentration and mixed-liquor volatile suspended solids (MLVSS) concentration were 6,260 ± 211 mg L^{-1} and 4,650 ± 175 mg L^{-1}, respectively. The influent was prepared based on the characteristics of Harbin region municipal wastewater, the composition of which was as follows: 350–400 mg L^{-1} COD (as sodium acetate), 50–60 mg L^{-1} NH_4^+–N (as ammonium chloride), 5–6 mg L^{-1} total phosphorus (TP) (as potassium dihydrogen phosphate) and 1.0 mL of trace solution per liter [20]. The pH value was maintained around 8 by 1 mol L^{-1} HCl and 1 mol L^{-1} NaOH.

2.2. Operational strategy

The experiment was carried out in a sequencing batch reactor (SBR). The reactor was fabricated of plexiglass with a working volume of 9.0 L and a volumetric exchange ratio of 50%. At the bottom of the reactor, an air diffuser and an inlet were used to introduce air and influent. The air was supplied by using an aerator with an airflow rate of 40.0 L min^{-1}. DO in the bulk liquid was automatically controlled during the aerobic reaction period. The whole granulation process is composed of lag, granulation and granule maturation phases [21]. The SBR was operated on a 4–6 h cycle. Each cycle involved 2 min of feeding, 30 min of stirring (110 rpm), 180 to 300 min of aerobic reaction (adjusted with the cycle experiment results), 5 to 15 of min settling, 2 min of discharge and the rest of the time was idle. During the experiment, the SBR operated at room temperature (20°C). The operational strategy is given in Table 1.

2.3. Analytical methods

COD, nitrogen compounds (NH_4^+–N, NO_2^-–N and NO_3^-–N), TP, suspended solids (SS), MLVSS, MLSS, sludge volume (SV) and sludge volume index (SVI) were analyzed at regular intervals according to the Standard Methods [22]. All tests were performed in triplicate. Statistical analysis was conducted by a *t*-test using SPSS software (SPSS 21.0). The DO and pH value were detected by DO Meter (Oxi 340i CellOx 325, WTW, Germany) and pH meter (muti 340i, WTW Company, Germany), respectively. The extraction of extracellular polymeric substances (EPS) was based on the formaldehyde and sodium hydroxide method [23], and the protein (PN) and polysaccharide (PS) contents were carried out according to the method reported by Badireddy et al. [24]. Average particle size and size distribution were measured by a laser particle size analysis system (Mastersizer 2000, Malvern instruments Ltd., UK). The granule morphology and the distribution of microorganisms were observed by an optical microscope (CX31, Olympus, Japan) and scanning electron microscope (SEM; JSM-5610LV, JEOL, Japan), respectively.

3. Results and discussion

3.1. Removal performance of the bioreactor

To promote aerobic granulation, the settling time was progressively reduced to 10 min during the lag phase (days 1–40) [25]. As shown in Fig. 1, The MLSS rapidly decreased from day 1 to 20. From day-20 onwards, the sludge loss stopped and MLSS stabilized around 2,100 mg L^{-1}. The effluent substrate concentrations were not stable in this phase because of the biomass washout. Subsequently, in the granulation phase (day-41–101), settling time was shortened to 5 min. The MLSS gradually increased and stabilized at ca. 6,000 mg L^{-1} towards the end of the granulation phase, which was strongly associated with the rapid growth of heterotrophic bacteria in this phase. As shown in Fig. 2, the effluent COD concentration was below 50 mg L^{-1} in most cycles of operation and removal efficiency was over 85% after 61 d of operation. The remaining COD probably

Table 1
Operational strategy of SBR

Parameters	Lag phase	Granulation phase	Granule maturation phase
Duration, (d)	1–40	41–101	102–141
DO set-point, (mg L^{-1})	1.0 ± 0.2	1.0 ± 0.2	1.0 ± 0.2
Aerobic phase duration, (min)	180–300	180	180
Settling time, (min)	10	5	5
Hydraulic retention time, (h)	12	8	8

owed non-biodegradable compounds such as soluble microbial products (SMPs). SMPs were derived from substrate metabolism and biomass decay [26]. Approximately 67.3% TP removal took place on the day-40 and the removal efficiency reached 90.6% on day-101, indicating that mature granule possessed excellent phosphorus removal capacity. The NH_4^+ and total nitrogen (TN) removal efficiencies increased gradually and finally stabilized at ca. 99% and 75%, respectively. In the granule maturation phase (days-102–141), the removal efficiencies of all nutrients mentioned above changed rarely. COD, NH_4^+, TN and TP removal was 91.0%, 98.6%, 77.6% and 92.5%, respectively, suggesting the aerobic granules system was successfully stabilized.

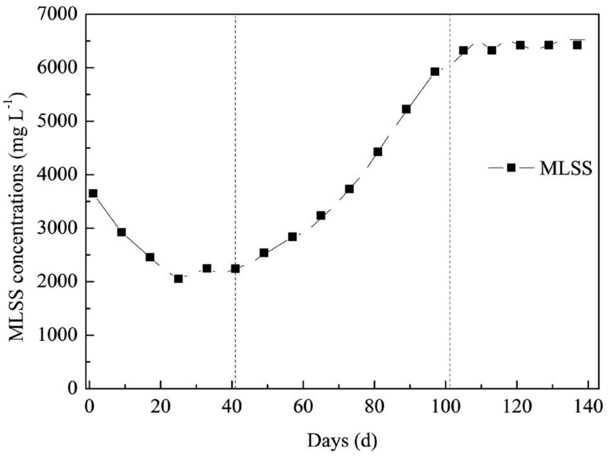

Fig. 1. Variations of MLSS of sludge in granulation during the experimental period of 140 d.

3.2. Reactor's performance throughout a cycle

Cycle studies were carried out to evaluate the feasibility and efficiency of aerobic granules process under low DO. As shown in Fig. 3, total COD removal of 29.1 mg L^{-1} occurred in the anaerobic phase. Phosphorus release of 2.09 mg L^{-1} could be attributed to PAOs metabolisms in the anaerobic phase, corresponding to carbon uptake of 14 mg L^{-1}. 15.1 mg L^{-1} COD was used as the carbon source in the denitrification process. NH_4^+ concentration showed a slight decrease which can be attributed to the adsorption capacity of the aerobic granules. As reported, ratios of carbon uptake to phosphorus release was 0.15 mg P/mg sodium acetate in the anaerobic metabolism of biological phosphate removal processes [27]

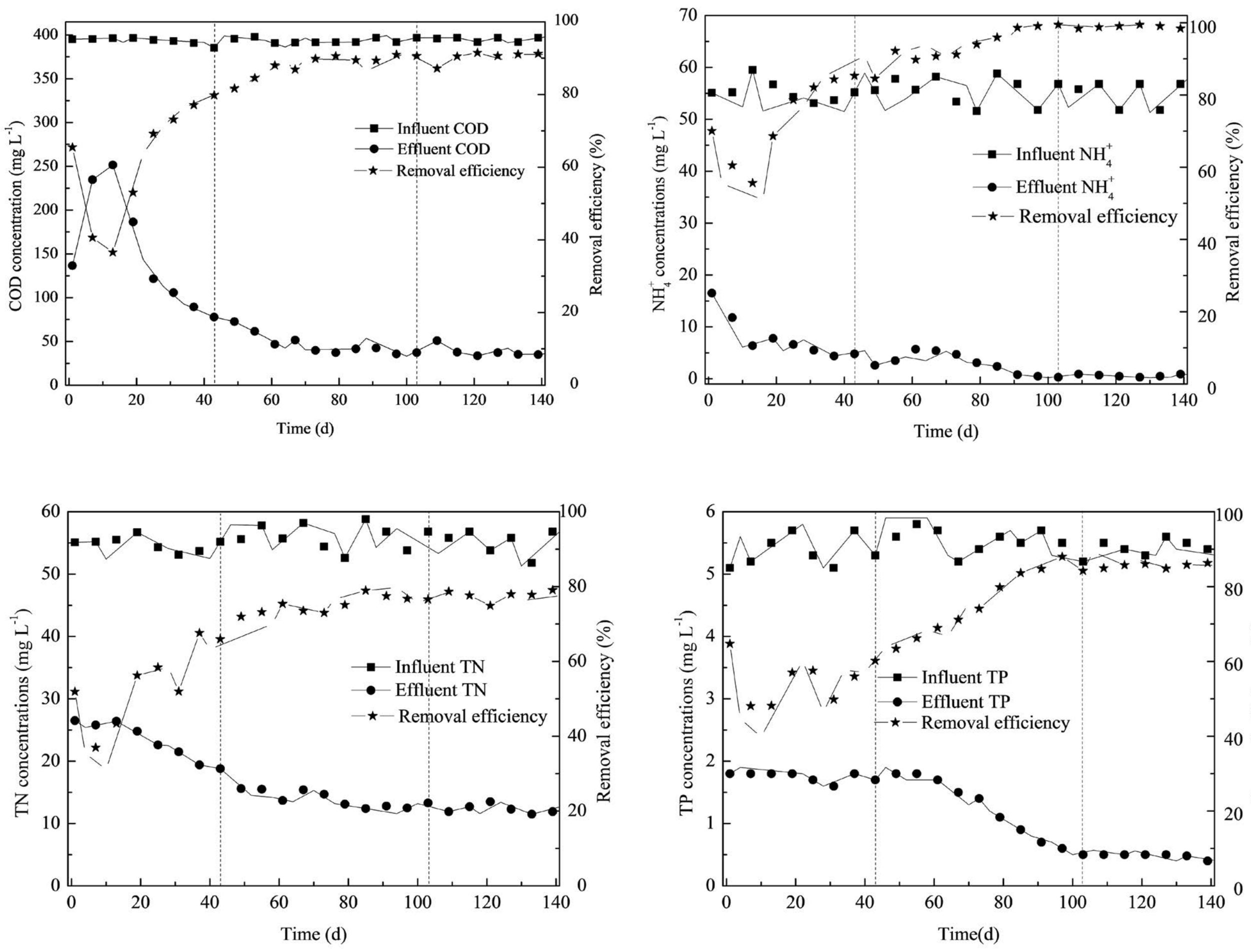

Fig. 2. The removal efficiency of nutrients during the cultivation process.

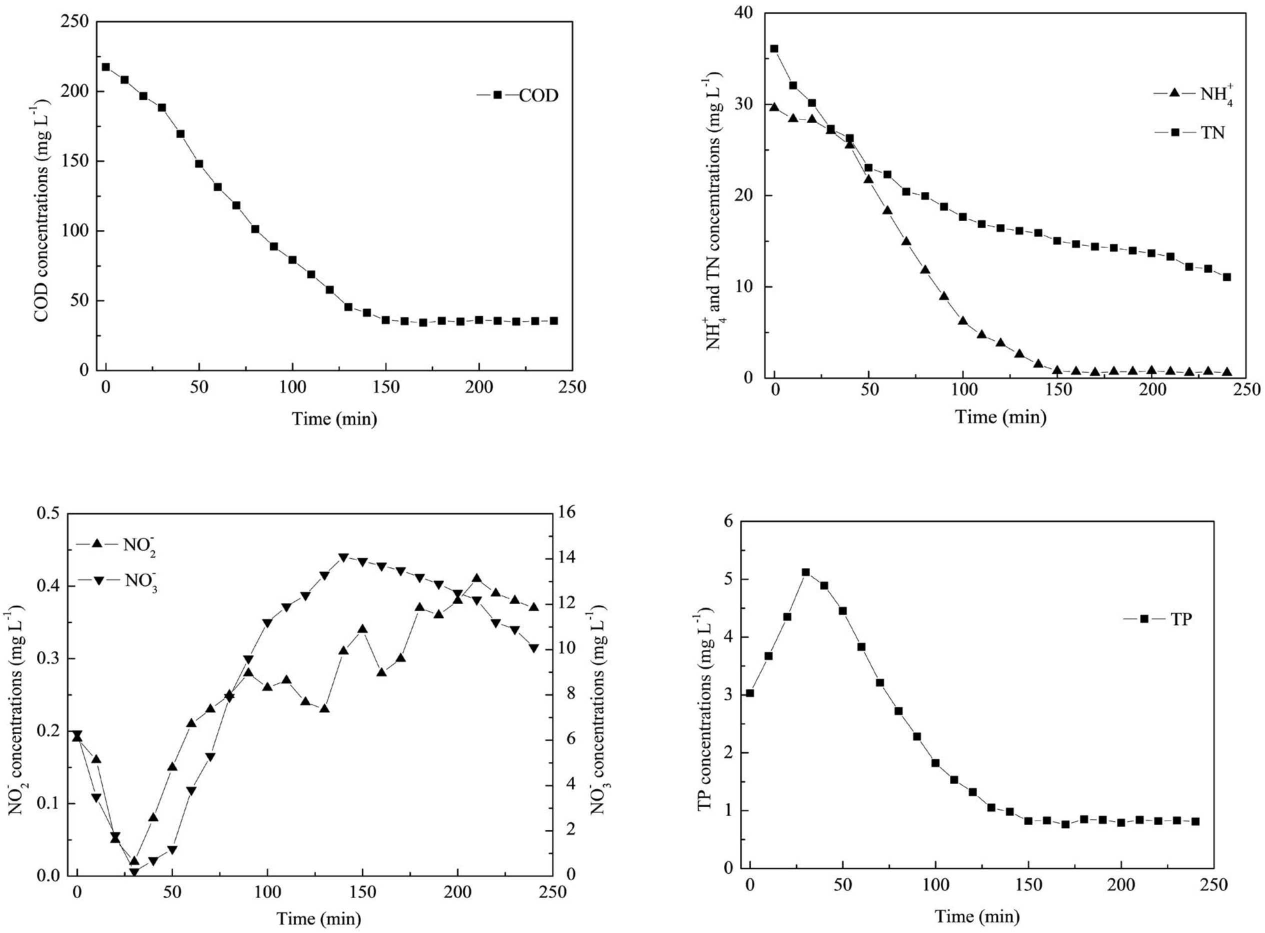

Fig. 3. Removal efficiencies of nutrients in a cycle.

and the COD/NO_3^- for denitrification of sodium acetate as an exotic additional carbon source was 3.66 mg sodium acetate/NO_3^- [28]. NH_4^+ was converted to NO_3^-, and phosphate was used by PAOs for polyphosphate formation in the aeration phase. At the beginning of the aeration phase, COD, NH_4^+ and TP concentrations sharply decreased, whereas TN concentration decreased in a step-wise manner. In this study, the NH_4^+ assimilation for cell growth accounted for ca. 4%. This is also supported by a study conducted by Wagner which demonstrated that the mature granules had a low ability to assimilate nitrogen in the aeration phase [25]. In this study, the simultaneous nitrification–denitrification (SND) efficiency was 52.2%, which is lower than that previously reported value (i.e., 60% [29,30]). SND in granules needs a certain thickness in the range of 200–7,000 μm [31,32]. A previous study suggested that an optimal granule size was in the range of 700–1,900 μm [33,34]. As shown in Fig. 4, the majority (56.2%) of the sludge had a granule size between 300 and 600 μm with an average granule size of 436.5 ± 12 μm. In this study, low DO had shown a positive effect on SND. In the settling, discharge and idle phases, NO_2^- production was barely detectable, with the average concentration in the bulk liquid of 0.37 ± 0.08 mg NO_2^- L^{-1}. A higher NO_3^- concentration was found. Probably this could be due to a lack of organic matter for denitrification.

3.3. Formation of aerobic granules

The changing pattern of sludge particle size throughout culture time was shown in Fig. 5. There was no obvious variance in sludge particle size during the lag phase. From day-41 onwards, the sludge particle size increased exponentially from 80.6 to 401.2 μm, named the granulation phase, and then stabilized at 436.5 μm on the day-141, which was lower than the previously reported values (2.5 mm [35] and 1.4 mm [36]). Small granules may reduce mass transfer resistance and improve good settleability [37]. SVI reflects the settling properties of the granules. The typical 5 min of sedimentation (SVI_5) value of <50 mL g^{-1} for aerobic granules was suggested by Cydzik-Kwiatkowska and Wojnowska-Baryła [38]. As shown in Fig. 6, from day-65 onwards, the SVI_5 value decreased below 50 mL g^{-1}, while small granules appeared in the reactor. The sludge was transformed into granules with a relatively loose structure. Liu and Tay [39] indicated that when the difference between SVI_5 and 30 min sedimentation (SVI_{30}) is lower than 10%, successful granulation was achieved. From day 97 onwards, the SVI_5/SVI_{30} ratio was nearly 1, indicating a completely granulated system. As shown in Figs. 7a and b, the matured granule exhibited a round shape with a dense and compact structure. Figs. 7c and d show that Filamentous

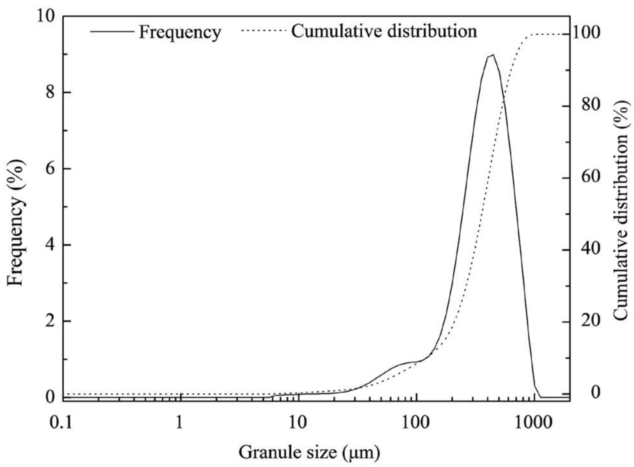

Fig. 4. Granule size distribution graph.

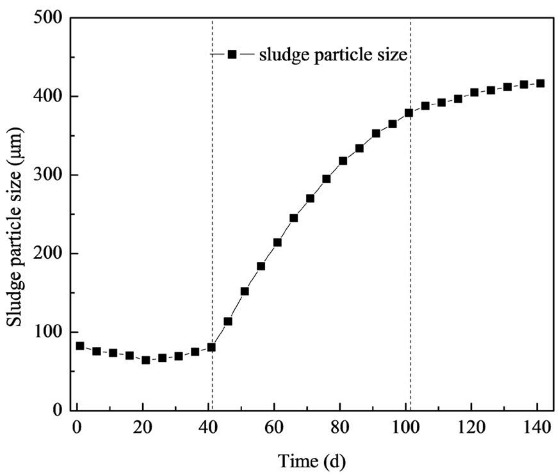

Fig. 5. The variation of steady particle size during the culture process.

bacteria on the surface seem to be embedded in EPS. These results agree well with the ones reported by Jiang et al. [40].

3.4. Variation of EPS

EPS are metabolic products, which mainly include polysaccharides, proteins, lipids and humic substances. The components in EPS could affect the physicochemical characteristics of the cellular surface. The contents of protein (PN), polysaccharide (PS) and protein to polysaccharide ratio (PN/PS) in EPS in the granulation phase and granule maturation phase are shown in Fig. 8. During the granulation phase, the polysaccharide level increased gradually to 30.6 mg g^{-1} VSS, whereas the protein level increased linearly up to almost 178.6 mg g^{-1} VSS. During the granule maturation phase, the polysaccharide level remained stable at 30.0 mg g^{-1} VSS, whereas the protein level slightly increased from 178.6 mg g^{-1} VSS on day 101 to 190.4 mg g^{-1} VSS on day 141. These phenomena indicated that the PN facilitated granulation, which was in line with published literature [40,41]. PN/PS ratio is a good indicator of the granule settleability and strength [42]. Generally, PN is the predominant component of the EPS [23,43]. This was consistent with our results. Other research concluded that PS was greater than PN [39,44]. The different results could result from the operating conditions. Many studies have shown that starvation time plays a role in the granulation process [1,39]. The starvation time in this study was 1 h which was shorter than those given by the above-mentioned authors (3–7 h). The study showed that the protein content increases in the proportion of the EPS components in the shorter starvation time while the polysaccharide components were relatively lower. When the starvation period was prolonged, EPS could be used as a substrate to maintain microbial survival. The utilization of protein was higher than that of polysaccharides, resulting in an upward trend in the PS/PN ratio of EPS [23].

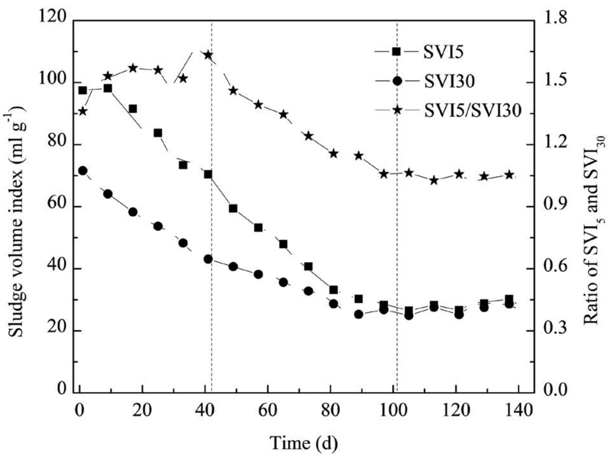

Fig. 6. Change of SVI at different cultural time.

3.5. Costs of aeration equipment

In practice, aeration rates corresponding to different DO were influenced by many factors (e.g., aeration equipment, characteristics of sewage, the structure of aeration tank, temperature, etc.) [45]. The formula of total air supply quantum calculation is shown as follows:

$$G_S = \frac{R \cdot C_{S(20)}}{\alpha\left[\beta \cdot \rho \cdot C_{\mathrm{Sb}(T)} - C\right] \times 1.024^{(T-20)} \times 0.3E_A} \cdot 100 \quad (1)$$

where G_S is the total air supply quantum, m^3 h^{-1}; R is the oxygen demand, kg h^{-1}; $C_{S(20)}$ is the saturation DO value of distilled water in 20°C, 9.17 mg L^{-1}; α is the correction coefficient, 0.85; β is the correction coefficient, 0.95; ρ is the correction coefficient of pressure, 1; $C_{\mathrm{Sb}(T)}$ is the saturation DO average value in T °C, mg L^{-1}; C is the DO value when aeration tank is in normal operation, mg L^{-1}; 1.024 is the temperature correction coefficient; T is the temperature, °C; E_A is the oxygen utilization coefficient, 10%.

The oxygen demand of wastewater in the actual condition is:

$$R = a'QS_r + b'VX_V \quad (2)$$

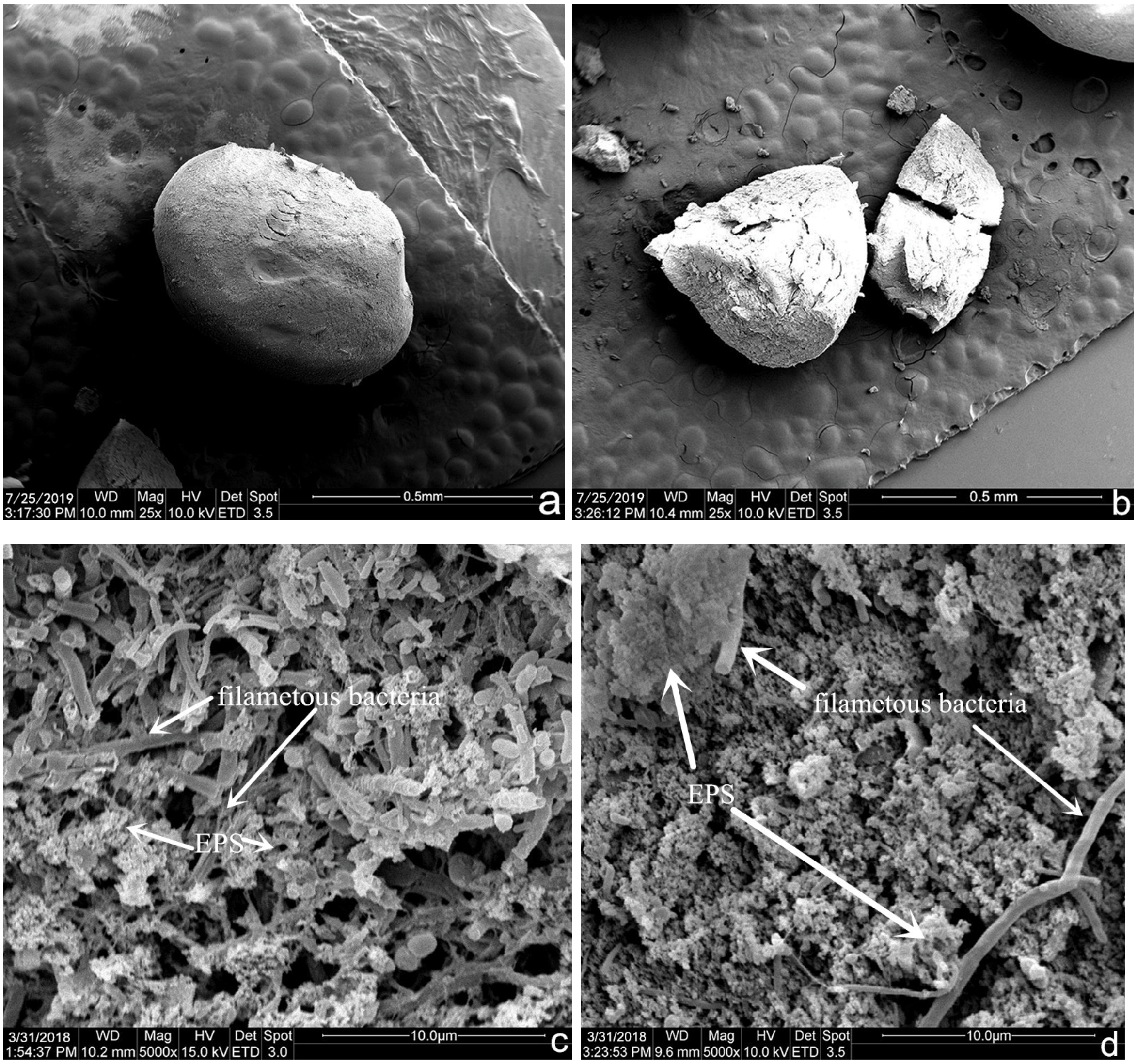

Fig. 7. Morphologies of mature granules.

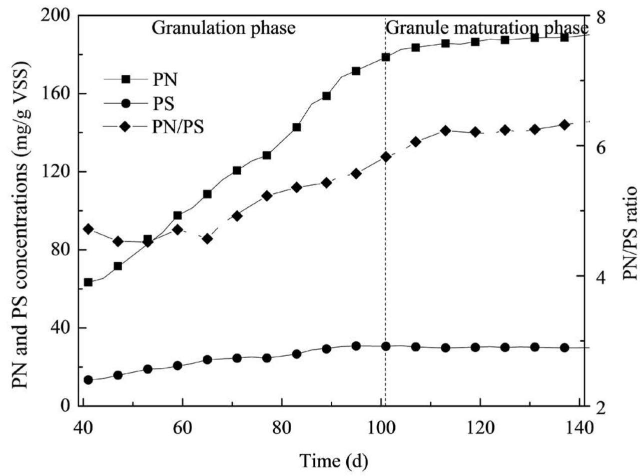

Fig. 8. EPS chemical components variations during the experiment.

where R is the oxygen demand of wastewater in actual condition, kg d^{-1}; a' is the oxygen demand rate for microorganism 0.42–0.53; Q is the flow, m^3 d^{-1}; S_r is the organic pollutant removal by a microorganism, kg BOD d^{-1}; b' is the oxygen demand of microbial endogenous respiration, 0.11–0.188; V is the volume of the reactor, m^3; X_V is the volatile suspended solid concentration, kg m^{-3}.

Saturation DO average value is:

$$C_{sb} = C_s\left(\frac{P_b}{2.026\times10^5} + \frac{21\left(1-E_A\right)}{42\left(79+21\left(1-E_A\right)\right)}\right) \quad (3)$$

$$C_{sb} = C_s\left(\frac{P_d}{2.026\times10^5} + \frac{21\left(1-E_A\right)}{42\left(79+21\left(1-E_A\right)\right)}\right) \quad (4)$$

where C_{sb} is the saturation DO average value, mg L^{-1}; C_s is the saturation DO value, mg L^{-1}; P_d is the blower outlet pressure, Pa.

Table 2
Variation of airflow rate under different DO (20°C)

DO (mg L^{-1})	Aeration rate (m^3 h^{-1})
1.0	2.12
1.5	2.47
2.0	2.94
2.5	3.65
3.0	4.79
3.5	6.97
4.0	12.79

Saturation DO average value:

$$C_{sb} = 9.17 \times \left(\frac{1.06 \times 10^5}{2.026 \times 10^5} + \frac{21(1-0.1)}{42(79+21(1-0.1))} \right) = 4.84 \text{ mg/L} \quad (5)$$

The oxygen demand of wastewater in actual condition:

$$R = 0.5 \times 0.018 \times 0.0043308 + 0.12 \times 9 \times 4.69 = 2.12 \times 10^4 \text{ kg/h} \quad (6)$$

Total air supply quantum:

$$G_s = \frac{2.12 \times 10^4 \times 9.17}{0.85 \times (0.95 \times 1 \times 4.84 - 1) \times 1.024^{20-20} \times 0.3 \times 0.1} \times 100 = 2.12 \text{m}^3/\text{d} \quad (7)$$

According to the test, aeration rates corresponding to different DO are listed in Table 2. As DO rise, aeration rates increase exponentially. At present, DO in the aerobic tank was generally controlled in 2–4 mg L^{-1}. Based on the actual system operating data from Taiping WWTP, DO in the aerobic tank was kept at 2.0 mg L^{-1}. In this study, when DO was controlled at 1.0 mg L^{-1}, aeration equipment produced less than a 27.9% aeration rate, compared with DO of 2.0 mg L^{-1}. Generally, the consumption for aeration was at least 0.18 kWh m^{-3} in WWTP [45]. For example, the amount of aeration equipment in Taiping WWTP was 8.2 × 10^7 m^3 a^{-1}, and the energy consumption of aeration equipment was 1.9 × 10^7 kWh a^{-1}. If aerobic granules technology was applied at DO of 1.0 mg L^{-1}, the energy consumption of aeration equipment could save 5.3 × 10^6 kWh a^{-1}, which equalled to 12% of the total power consumption.

4. Conclusion

The formation of aerobic granules was achieved by using low-strength wastewater under low DO conditions. The matured granule exhibited a round shape with a dense and compact structure, and the average granule size was 436.5 ± 12 μm. COD, NH_4^+, TN and TP removal in the granulation phase reached to 91.0%, 98.6%, 77.6% and 92.5%, respectively. The aerobic granule technology at low DO could save 12% of the total power consumption of WWTP. Further studies will be necessary to validate these results in a full-scale reactor treating real wastewater.

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