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Research article

Coupling high-rate activated sludge process with aerobic granular sludge process for sustainable municipal wastewater treatment

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ABSTRACT

Achieving a neutral/positive energy balance without compromising discharge standards is one of the main goals of wastewater treatment plants (WWTPs) in terms of sustainability. Aerobic granular sludge (AGS) technology promises high treatment performance with low energy and footprint requirement. In this study, high-rate activated sludge (HRAS) process was coupled to AGS process as an energy-efficient pre-treatment option in order to increase energy recovery from municipal wastewater and decrease the particulate matter load of AGS process. Three different feeding strategies were applied throughout the study. AGS system was fed with raw municipal wastewater, with the effluent of HRAS process, and with the mixture of the effluent of HRAS process and raw municipal wastewater at Stage 1, Stage 2 and Stage 3, respectively. Total suspended solids (TSS), chemical oxygen demand (COD), ammonia nitrogen (NH_4^+-N), and total phosphorus (TP) concentrations in the effluent were less than 10 mg/L, 60 mg/L, 0.4 mg/L, and 1.3 mg/L respectively at all stages. Fluctuations were observed in the denitrification performance due to changes in the influent COD/total nitrogen (TN) ratio. This study showed that coupling HRAS process with AGS process by feeding the AGS process with the mixture of HRAS process effluent and raw municipal wastewater could be an appropriate option for both increasing the energy recovery potential of WWTPs and enabling high effluent quality.

1. Introduction

Upgrading wastewater treatment plants (WWTPs) and/or applying novel wastewater treatment technologies has been essential to meet stringent discharge standards (Wang et al., 2012). Land and energy requirements together with excess sludge handling cost are the key factors that need to be considered solidly to obtain a feasible treatment in WWTPs in terms of technical and economic aspects (Chen et al., 2020; Zaborowska et al., 2021). Nowadays, efficient use of resources has become vital and thus, efforts on sustainability have been put into practice for WWTPs. Since energy is a crucial element of sustainability, achieving a neutral/positive energy balance in WWTPs without compromising discharge standards becomes to be one of the main targets (Maktabifard et al., 2020). There are two ways for achieving sustainability in WWTPs in terms of energy: Decreasing energy demand in WWTPs and maximizing energy harvest from the wastewater (Maktabifard et al., 2018; Yan et al., 2020).

Biological nutrient removal (BNR) plants generally consist of suspended growth bioreactors having different redox zones, and discrete secondary clarifier tanks, where solids are separated and recirculated to the bioreactors. Various modifications have been developed for the BNR processes in order to increase the nutrient removal efficiency. Various activated sludge (AS) process configurations such as A^2/O, modified (five-stage) Bardenpho, and modified UCT have been applied for biological wastewater treatment technology to remove carbonaceous matter and nutrients (nitrogen, phosphorus) over the century (Jenkins and Wanner, 2014). However, large footprint and energy requirement of nutrient removing AS processes accelerated the research for new

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technologies (Ferreira et al., 2021).

Aerobic granular sludge (AGS) process offers up to 75% decrease in footprint requirement and 30–50% less energy consumption compared to A^2/O process (Pronk et al., 2015; Thwaites et al., 2018). In the AGS process, aerobic granules are responsible for the biological removal in contrast to flocs in the AS processes. There are different redox layers inside the granules that create optimum conditions for the removal of carbon and nutrient in a single reactor (de Kreuk et al., 2007; Adav et al., 2008). The inner anoxic/anaerobic zone is occupied by denitrifiers and facultative phosphate accumulating organisms (PAOs) (Winkler et al., 2015), while the surface aerobic zone is dominated by aerobic heterotrophs and autotrophs, mainly nitrifiers (Liu, 2006; Winkler et al., 2015). Therefore, simultaneous carbon, nitrogen, and phosphorus removal can be achieved in a single granule (de Kreuk et al., 2005).

Presently, there are more than 80 full-scale AGS treatment plants operated in the world, and it is seen as one of the key technologies of wastewater treatment for next century (van Loosdrecht and Brdjanovic, 2014; van Haandel and van der LubbeDesign, 2015; Ferreira et al., 2021; Hamza et al., 2022). In full-scale AGS applications for municipal wastewater treatment, effluent chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) concentrations have been reported between 64 and 90 mg/L, 6.9–10 mg/L, and 0.9–3.2 mg/L respectively (Li et al., 2014; Pronk et al., 2015). Although AGS process promises high treatment performance, transforming the activated sludge flocs into granules takes long time (Kosar et al., 2022; van Dijk et al., 2022). AGS process can be combined as a pre-treatment or post-treatment alternative to different wastewater treatment configurations to either decrease the granulation time or increase the treatment performance (Giesen and Thompson, 2013; Pronk et al., 2017). Another important issue regarding AGS systems is the excess granular sludge. The excess sludge from AGS process can be used as inoculum to accelerate aerobic granulation in other reactors, which are in start-up period (Li et al., 2015a; Long et al., 2014). In the scope of the circular economy concept, phosphorus, biopolymers (Kaumera) (including alginate-like exopolysaccharides (ALE)), and polyhydroxyalkanoates (PHA) in AGS sludge can be extracted and evaluated for beneficial use (Ferreira et al., 2021). The other important value of the excess sludge is its energy content. Energy can be recovered by conversion of organic content of excess biological sludge to methane via anaerobic digestion. However, excess biological sludge has less energy content compared to primary sludge. Because most of the organic content in wastewater is used by biomass in the bioreactors and converted to CO_2, thus, less organic content is available in the excess sludge. Carbon capture processes are gaining popularity to maximize energy recovery from wastewater (Rahman et al., 2020). In carbon capture processes, carbon in the wastewater is redirected to anaerobic digesters through the waste sludge without losing it to the atmosphere in CO_2 form. The integration of carbon capture processes with the AGS process can be attractive for maintaining energy balance and sustainability in AGS plants.

High-rate activated sludge (HRAS) process as a carbon capture process is used as the first stage of two-step AS system, represented as adsorption bio-oxidation (AB) process (Bohnke, 1983). HRAS process has been getting attention due to an increasing tendency to the concept of carbon capture from wastewater in recent years. Carbon loss via mineralization is reduced with low sludge retention time and low dissolved oxygen (DO) concentration in HRAS process, and concentrated carbon in the excess sludge can be anaerobically digested for energy production. Lack of nitrification in the HRAS process results in high concentrations TN in the effluent (Guven et al., 2019). For this reason, HRAS system, represented as high loaded biological adsorption stage (A Stage), is applied as the first step of AB process, while nutrient is removed by the second step, referred as low loaded bio-oxidation stage (B stage) (de Graaff et al., 2016; Wan et al., 2016). Around 60% of the influent COD could be removed by HRAS process and the effluent has a low COD/TN ratio, so it is usually integrated with low COD needed treatment processes, e.g. shortcut biological nitrogen removal process (SBNR) and partial nitrification with anammox process (Wan et al., 2016; Guven et al., 2017; Rahman et al., 2019).

This study aims to combine the energy recovery potential of HRAS process with the high treatment performance of AGS process by feeding HRAS process effluent to AGS system. In this scope, AGS process was fed with raw municipal wastewater; effluent of an HRAS process; and mixture of the effluent of HRAS process and raw municipal wastewater, in order. Sludge characteristics and treatment performance were studied comprehensively in the AGS process. Coupling of both processes can improve the potential for energy recovery from wastewater while obtaining good effluent quality, thus sustainable wastewater treatment can be achieved. For this purpose, the effect of coupling of the two processes on the treatment performance was studied, and an appropriate treatment configuration was proposed.

2. Material and methods

2.1. Experimental set-up

A laboratory-scale AGS reactor (plexiglass, 180 cm of height, 10 cm of diameter) with a specially made rubber diffuser, and online sensors (pH (Kuntze Instrument, Germany), temperature (Meter, Turkey), DO (Aqualabo, France), oxidation reduction potential (ORP) (Kuntze Instrument, Germany), and level sensor (IFM, Germany) was used in the study (Fig. 1). Wastewater in the influent tank was fed from the bottom point of the reactor via feed pump (Seepex, Germany), while treated wastewater was transferred to the effluent tank via discharge pump (Lead Fluid, China). The influent flowrate was monitored via a flowmeter (IFM, Germany). During anaerobic phase, recirculation from the top of the reactor to the bottom was performed via recirculation pump (Lead Fluid, China).

2.2. Seed sludge and wastewater characterization

The laboratory-scale AGS system was seeded with waste activated sludge (WAS) taken from a full-scale municipal wastewater treatment plant operated as a BNR system. Characterization of the seed sludge is given in Table 1. The AGS system was fed at three stages with different influents including (i) raw municipal wastewater (Stage 1), (ii) effluent of HRAS process treating the same raw municipal wastewater (Stage 2), iii) mixture of raw municipal wastewater (20% of the mixture) and effluent of HRAS process (80% of the mixture) (Stage 3). Characterization of the influent for each stage is given in Table 2. Raw wastewater (Stage 1) had the highest COD/TN ratio, whereas the effluent of HRAS process (Stage 2) had the lowest COD/TN ratio.

2.3. Experimental procedure

The study consisted of three stages based on influent type. The AGS reactor was operated on simultaneously fill/draw sequencing batch reactor (SBR) mode. Six cycles per day were applied in SBR mode. Duration of each cycle was 240 min including fill/draw phase (30 min), anaerobic phase (50 min), aerobic phase (128 min), settling phase (30 min), and idle phase (2 min). Height to diameter (H/D) ratio of the AGS reactor was 15 (water level was 150 cm in the reactor) and volume exchange ratio (VER) was provided as 50%. The AGS system was inside an air-conditioned laboratory, so the temperature was almost stable during the operation period. pH arrangement was not needed during operation. Due to the change in the influent characterization among three stages, OLR has also changed at each stage. OLR was 1.24 ± 0.03 kg COD/m^3. day, 0.46 ± 0.03 kg COD/m^3. day, and 0.77 ± 0.03 kg COD/m^3. day at Stage 1, Stage 2 and Stage 3, in order.

2.4. Analytical methods

TSS, VSS, COD, TN, NH_4^+-N, and TP measurements were conducted

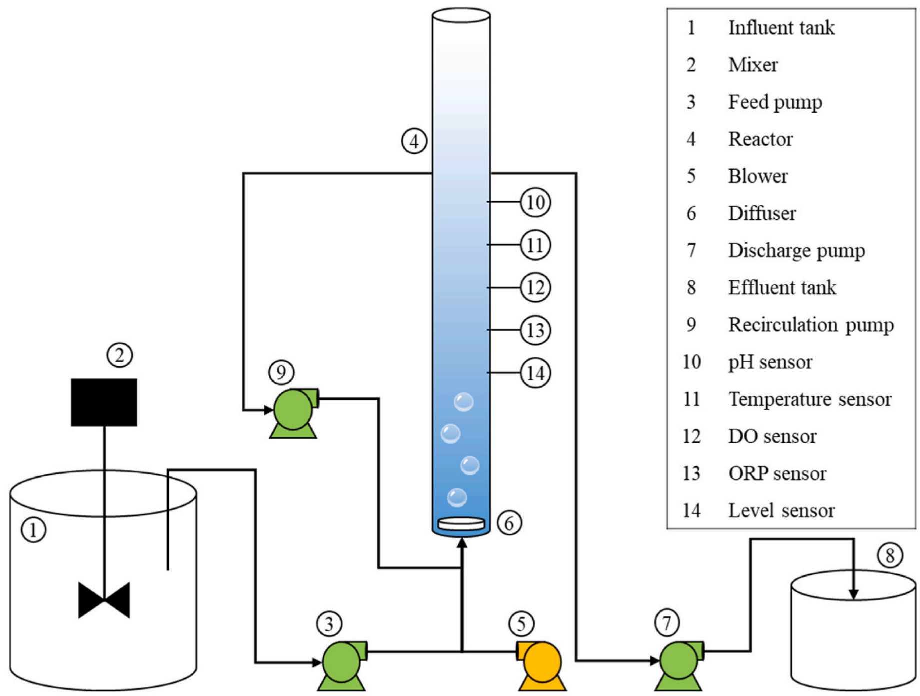

Fig. 1. Experimental set-up of lab-scale AGS system.

Table 1
Characterization of the seed sludge.

Parameter	Unit	Value (Average ± Standard Deviation (SD))
COD	mg/L	15,462 ± 340
Soluble COD (sCOD)	mg/L	169 ± 16
sCOD/COD	%	1.1 ± 0.04
Total Suspended Solids (TSS)	mg/L	11,460 ± 220
Volatile Suspended Solids (VSS)	mg/L	8110 ± 230
Sludge Volume Index$_5$ (SVI_5)	mL/g	154 ± 5
SVI_{30}	mL/g	78 ± 1.0
Normalized Capillary Suction Time (CST)	sec/g TSS/L	1.44 ± 0.09
pH	–	7.30 ± 0.02
Median Particle Size (d_{50})	μm	49 ± 0.4

Table 2
Characterization of the influent to the AGS reactor.

Parameter	Unit	Value (Average ± SD)		
		Stage 1	Stage 2	Stage 3
COD	mg/L	414 ± 12	153 ± 11	257 ± 11
sCOD	mg/L	209 ± 9	100 ± 10	139 ± 7
TSS	mg/L	307 ± 12	46 ± 6	61 ± 3
TN	mg/L	51 ± 2	43 ± 4	44 ± 4
Ammonium Nitrogen (NH_4^+-N)	mg/L	35 ± 1	34 ± 4	35 ± 2
TP	mg/L	6.0 ± 0.4	2.6 ± 0.2	3.2 ± 0.1
Turbidity	NTU	217 ± 7	37 ± 2	59 ± 4
pH	–	7.47 ± 0.04	7.51 ± 0.08	7.51 ± 0.02
COD/TN	–	8.2 ± 0.5	3.6 ± 0.3	5.9 ± 0.6

according to Standard Methods (APHA, 2017). Samples were filtrated through 0.45 μm filters prior to soluble COD (sCOD) measurement. Turbidity and particle size distribution (PSD) measurements in the influent and effluent samples were conducted by a turbidimeter (Hach 2100 P, USA) and a PSD analyzer (Zetasizer Nano-S, Malvern Instrument, UK), respectively. Ion chromatography system (Dionex ICS – 300) was used for nitrate (NO_3^-) measurements. PSD of the sludge was measured by using Mastersizer 2000(Malvern Instruments, Hydro 2000 MU, UK). SVI_{30} of the sludge was determined according to Standard Methods (APHA, 2017). The volumes of 1 g of sludge after 5 min and 10 min of settling time were measured as SVI_5 and SVI_{10}, respectively. Batch settling tests were applied to measure hindered settling velocities of the sludge samples (van Loosdrecht et al., 2016). CST of the sludge samples was measured with 18 mm funnel by a CST analyzer (Triton Electronics, Type 304 M, UK).

Environmental scanning electron microscopy (ESEM) (Thermo Fisher Scientific, FEI Quanta FEG 250, USA) was used to observe surface morphology of the dried granules. To increase the conductivity of the samples, palladium (Pd) and gold (Au) were used in coating the samples with a vacuum evaporator (Quorum SC7620, UK) prior to ESEM analysis. Organics on the surface of the dried granules were determined by using Fourier transform infrared spectroscopy (FTIR) (PerkinElmer, Spectrum 100, USA). The method including high temperature-sodium carbonate (Na_2CO_3) extraction, that was described by Felz et al. (2016), was used for the extraction of extracellular polymeric substances (EPS). Extracellular proteins (pEPS) and extracellular carbohydrates (cEPS) were determined using bovine serum albumin and d-glucose monohydrate as standards, respectively. Modified Lowry method was used for protein measurements (Frolund et al., 1995). For carbohydrate measurements, the method described in Dubois et al. (1956) was used.

Effluent quality at each stage was compared using effluent quality index (EQI) Equation (1)) (Copp, 2002). EQI was calculated using the average concentrations of TSS, COD, nitrate and nitrite (NO), Total Kjeldahl nitrogen (TKN), and TP in the effluent. The weighting factors for different parameters were used as $\beta_{TSS} = 2$, $\beta_{COD} = 1$, $\beta_{NO} = 1$, $\beta_{TKN} = 20$, and $\beta_{TP} = 20$. β_{TSS}, β_{COD}, β_{NO}, and β_{TKN} were selected as suggested by Copp (2002). Weighting factor for TP parameter was selected same as the one used for TKN parameter (Gernaey and Jorgensen, 2004).

$$\begin{aligned} EQI = {} & \left(\beta_{TSS} \times TSS_{effluent}\right) + \left(\beta_{COD} \times COD_{effluent}\right) + \left(\beta_{NO} \times NO_{effluent}\right) \\ & + \left(\beta_{TKN} \times TKN_{effluent}\right) + \left(\beta_{TP} \times TP_{effluent}\right) \end{aligned} \tag{1}$$

3. Results and discussion

3.1. Treatment performance

After seeding the system with flocculent sludge and feeding it with raw wastewater, the AGS process reached steady-state conditions on the 61st day, then, TSS concentration in the reactor and the pollutant concentrations in the effluent became stable. Then the AGS reactor was fed with raw wastewater in stage 1, the effluent of HRAS process, and a mixture of the effluent of HRAS process and raw wastewater in stage 3. Average effluent TSS concentration was lower than 10 mg/L at each stage. The highest TSS removal efficiency was obtained as 97 ± 0.4% at Stage 1. At Stage 2 and Stage 3, average TSS removal efficiencies were 85 ± 3% and 87 ± 2%, respectively. The highest COD removal efficiency was also achieved at Stage 1 (COD removal efficiencies: 86 ± 1.1% (Stage 1), 71 ± 2.7% (Stage 2), 84 ± 1.9% (Stage 3)). Average concentrations of COD, TP, TN, and NH_4^+-N parameters in the effluent were 58.5 ± 3.6 mg/L, 1.28 ± 0.17 mg/L, 12.5 ± 1.4 mg/L, and 0.35 ± 0.16 mg/L, respectively at Stage 1 (Fig. 2). At Stage 2, in which the effluent of HRAS process was fed to the AGS system, COD, TP, and NH_4^+-N concentrations in the effluent were decreased to 43.3 ± 2.1 mg/L, 1.08 ± 0.08 mg/L, and 0.29 ± 0.09 mg/L, respectively. However, higher concentration of TN (17.9 ± 0.9 mg/L in average) was observed in the effluent at Stage 2. Lower COD/TN ratio in the influent at Stage 2 (3.6 ± 0.3) compared to Stage 1 (8.2 ± 0.5) hampered denitrification, and thus decreased the nitrogen removal efficiency of AGS process. Limited denitrification capacity resulted in an increase in the effluent nitrate nitrogen (NO_3–N) concentration at Stage 2 in comparison to Stage 1, from 8.8 ± 0.6 mg NO_3–N/L to 11.4 ± 0.7 mg NO_3–N/L. At Stage 3, effluent of a pilot-scale HRAS process was mixed with raw municipal wastewater to increase the COD/TN ratio of the influent from 3.6 ± 0.3 to 5.9 ± 0.6. As a result, average TN and NO_3–N concentrations in the effluent decreased to 10.8 ± 1.8 mg/L and 7.4 ± 0.6 mg/L, respectively. Average COD, TP, and NH_4^+-N concentrations in the effluent were 39.8 ± 4.2 mg/L, 1.14 ± 0.18 mg/L, and 0.25 ± 0.14 mg/L, respectively at Stage 3 (Fig. 2). Effluent COD and TP concentrations were consistent with the data reported for full-scale AGS processes in the literature (COD: 40–90 mg/L, TP: 0.9–3.2 mg/L) (Giesen and Thompson, 2013; Pronk et al., 2015). At Stage 1 and Stage 2 similar average d_{50} values (415 ± 21 nm and 443 ± 59 nm, respectively) in the effluent were obtained. On the other hand, average d_{50} value in the effluent decreased to 263 ± 10 nm at Stage 3 (Fig. 3).

3.2. Sludge characteristics

TSS concentration in AGS reactor at the first day of operation was 6735 mg/L, it decreased to 4521 mg/L at Day 19 because of sludge washout from the system. After Day 20, TSS concentration started to increase, and once the system reached to steady state condition at Day 61, stable TSS concentrations (in the range of 5295–6484 mg/L) were measured in the AGS process. VSS/TSS ratio changed in the range of 77–84% during the whole study.

The values of different parameters representing sludge characteristics are given in Fig. 5 D_{50} values of the sludge at Stage 1 (82 ± 2 μm)

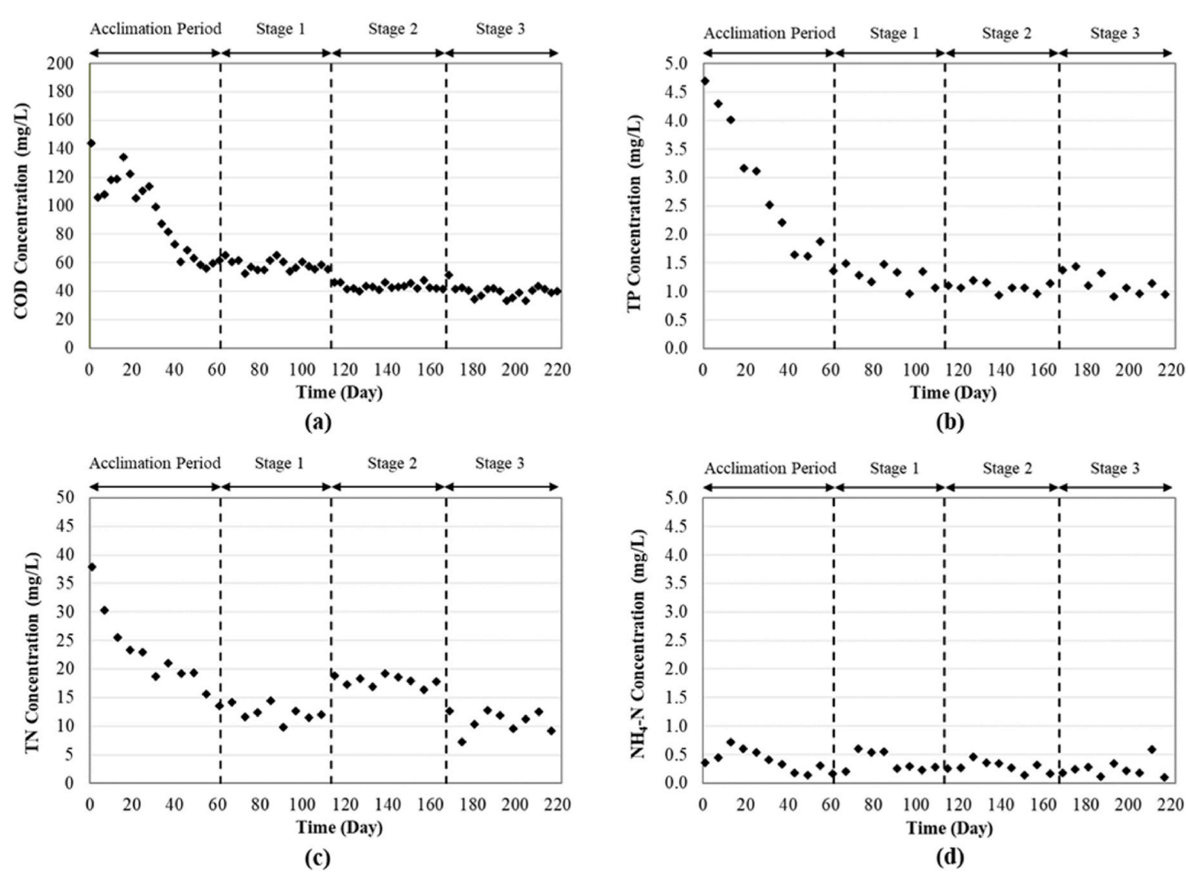

Fig. 2. The concentrations of different parameters in the effluent of AGS system: (a) COD, (b) TP, (c) TN, (d) NH_4^+-N.

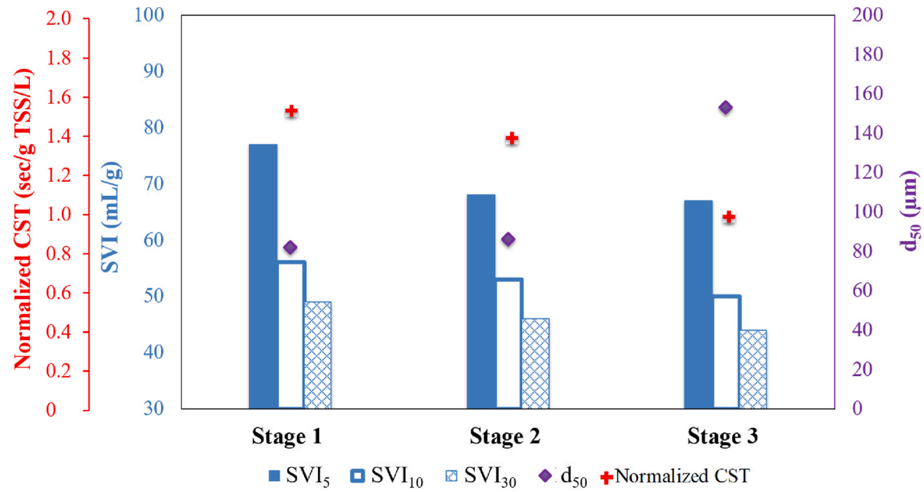

Fig. 3. SVI, d_{50}, and normalized CST values of the sludge in AGS process.

and Stage 2 (87 ± 3 μm) were similar, whereas higher d_{50} value (153 ± 11 μm) was obtained at Stage 3. Aerobic granules are formed when microorganisms come together and make a cluster on top of each other. The size of the granules is a significant parameter for the indication of nitrogen removal efficiency (Bathe et al., 2005). Small granules allow oxygen to reach inner layers, while big granules can contain anoxic/anaerobic zones to complete denitrification (Nguyen Quoc et al., 2021). In this study, the effluent nitrate concentration increased from 8.8 ± 0.6 mg/L at Stage 1–11.4 ± 0.7 mg/L at Stage 2, since the lowest influent COD/TN ratio was at Stage 2 (3.6 ± 0.3). The effluent nitrate concentration decreased to 7.4 ± 0.6 mg/L at Stage 3. Increase in COD/TN ratio of the influent from 3.6 (Stage 2) to 5.9 (Stage 3) resulted in an increase in granule size as well as denitrification capacity. Without sufficient available carbon in the influent, the ratio of nitrifiers to heterotrophs may increase, moreover, denitrifiers were dominated by ordinary aerobic heterotrophs. Thus, the absence of sufficient anoxic/anaerobic volume in the core of granules might restrict the enlargement of granules. This might be the reason for obtaining smaller sized granules at Stage 2 compared to Stage 3.

At Stage 2 and Stage 3, similar SVI_{30} values were observed, while at Stage 1 SVI_{30} values was slightly higher. At all stages, SVI_{30} values was compatible with the values (30–67 mL/g) reported in the literature for AGS process treating municipal wastewater (Bengtsson et al., 2018). Similar SVI_{30}/SVI_{10} ratios were obtained at Stage 1, Stage 2, and Stage 3 (0.86 ± 0.01, 0.86 ± 0.02, and 0.89 ± 0.09, respectively). Compared to the SVI_{30}/SVI_{10} ratio of seed sludge (0.51 ± 0.02), it can be interpreted that granulation was achieved at all stages. The best normalized CST value (0.99 ± 0.05 s/g TSS/L) was achieved at Stage 3. Zhang et al. (2020) reported a normalized CST value of 0.88 ± 0.01 s/g TSS/L for an AGS process treating domestic wastewater, which is compatible with the findings in our study.

Another test, that can be used to evaluate the settleability of the sludge, is hindered settling velocity measured by batch settling test. The fastest settling velocity was achieved at Stage 3 (6.3 m/h), while at Stage 1 (1.05 m/h), the granular sludge has the slowest settling characteristics. The settling velocity was measured at Stage 2 as 4.2 m/h. This finding was also consistent with the SVI values obtained at each stage.

ESEM images of the granule surfaces at each stage are given in Fig. 4. Porous structures were observed on the surface of the granules at each stage. Granules observed at Stage 1 and Stage 3 had more amorphous structures compared to Stage 2. At Stage 2, AGS process was fed solely with the effluent of HRAS process, which had almost six times lower TSS concentration than raw wastewater (Table 2). The absence of particulate matter inside the feed could be the reason for smoother granule surface observed at Stage 2. Inorganic particulate structures in the raw wastewater that attached to granules at Stage 1 could be seen on the surface of the granules Fig. 4(a).

Similar peaks were recorded for the FTIR spectrum of the granule surfaces at each stage (Fig. 5). At Stage 2 and Stage 3, stronger peaks were observed at 3276 cm^{-1} and 1636 cm^{-1} indicating polysaccharide and protein structures, respectively (Li et al., 2015b; Isik et al., 2019). The broad region of transmittance between 3600 and 3000 cm^{-1} might be caused by O–H stretching of hydroxyl functional groups (Kumar et al., 2006). C–H stretching was probably responsible for peaks at 2925 cm^{-1} at each stage (Bramhachari et al., 2007). Peaks at 3276 cm^{-1} (O–H stretching), 2925 cm^{-1} (weak C–H stretching), and 1636 cm^{-1} (asymmetric stretching of O–C–O) indicated EPS structures in the granules (Lin et al., 2010, 2013). Weak peaks at 1240 cm^{-1} (attributed to nucleic acids due to a band of phosphate group absorption) might also be caused by EPS presence in the granules (Sheng et al., 2006; Lin et al., 2010). Strong peaks at 1029 cm^{-1} (C–OH stretching) represented polysaccharides (Allen et al., 2004; Low et al., 2021).

EPS is considered as a material acting like gelatin, which functions as a network for microorganisms to grow (Flemming et al., 2007). Therefore, the interaction between EPS and microbial cells affects the formation of granular sludge (Ni, 2013). FTIR results indicated EPS presence at all stages. pEPS and cEPS measurements are given in Table 3. The highest pEPS formation was measured at Stage 3. At Stage 1 and Stage 2, similar values have been obtained. cEPS amounts were close to each other at all stages. Therefore, pEPS/cEPS ratio of granules was different at each stage. A higher pEPS/cEPS ratio indicates better granulation (Zhang et al., 2017). Consistent with the literature, it was observed that the pEPS/cEPS ratio correlated with granule size (p: 0.01; r: 0.93), and the highest pEPS/cEPS ratio observed at Stage 3 (d_{50} value: 153 ± 11 μm).

3.3. Overall evaluation

In this study, AGS system was fed with influent with different characteristics in each stage. That is why instead of pollutant removal efficiencies, EQI was used to compare the treatment performance of different stages. EQI was calculated by using the selected specific weighting factors for the concentrations of TSS, COD, NO_3–N, TKN, and TP parameters in the effluent at each stage. EQI was found as 6.6 g pollution/d, 7.8 g pollution/d and 5.4 g pollution/d at Stage 1, Stage 2 and Stage 3, respectively. Fig. 6 compares different stages in terms of EQI and sludge characteristics (d_{50}, pEPS/cEPS, and normalized CST values). The best effluent quality, with the highest d_{50} value and pEPS/cEPS ratio of the sludge, was obtained in Stage 3. Higher pEPS/cEPS ratio (Table 3) could be the reason of increased granule size. At Stage 1

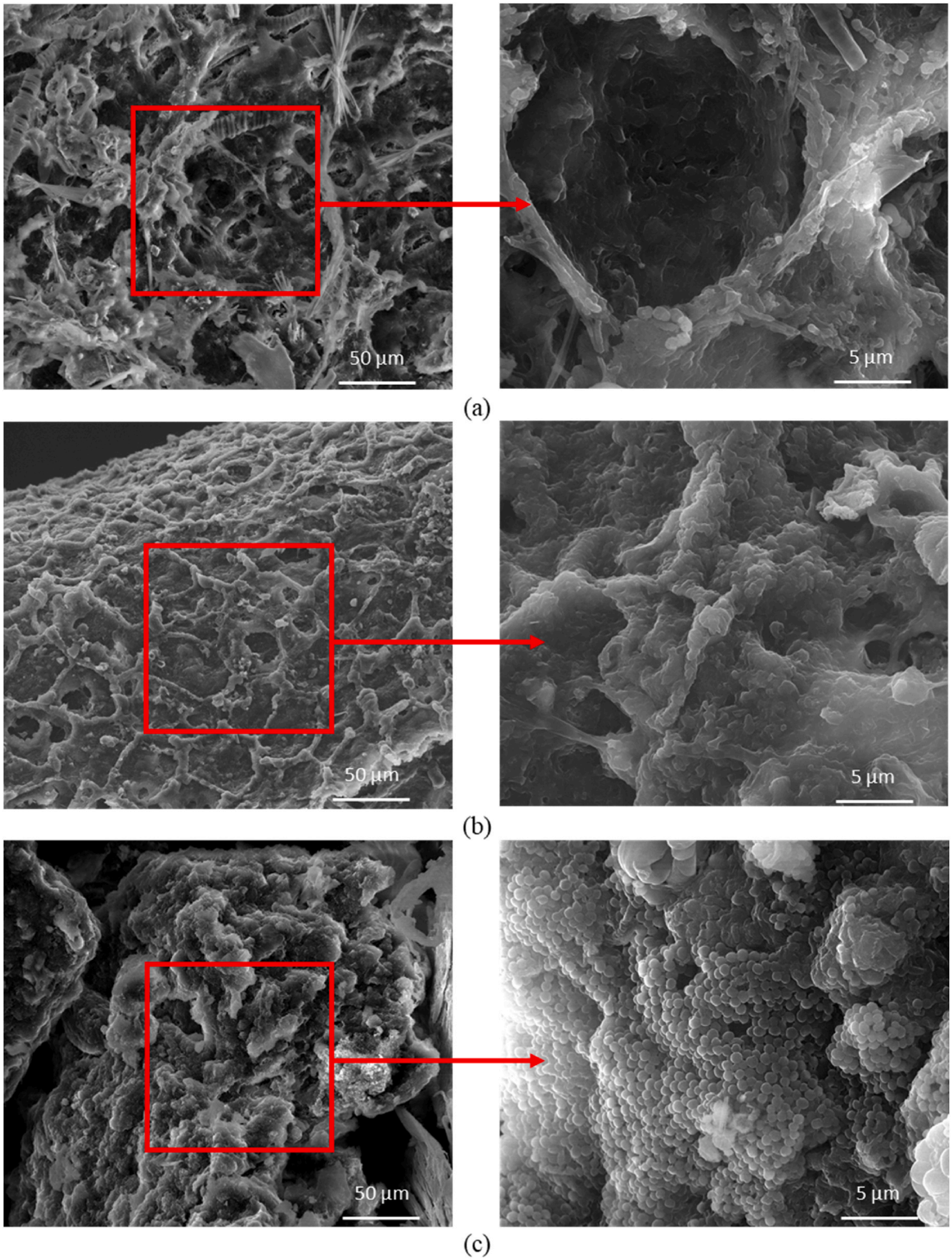

Fig. 4. ESEM images of granules: (a) Stage 1, (b) Stage 2, (c) Stage 3.

and Stage 2, similar pEPS/cEPS ratios correlated with similar d_{50} values. The lowest normalized CST, that was observed at Stage 3, would reduce chemical consumption in sludge handling facilities. The worst effluent quality was observed at Stage 2. The lowest influent COD/TN ratio might cause a decrease in the denitrification capacity of the granules at Stage 2. Considering overall evaluation, influent COD/TN ratio was found to be an important parameter that affects nitrogen removal in AGS process. In this study, the HRAS process effluent and raw wastewater were mixed to reduce the inlet pollutant load for the AGS process, while providing sufficient carbon for the denitrifiers and PAOs, thus balancing between capturing the carbon in the wastewater via HRAS process and obtaining high quality effluent in the AGS system was shown to be achievable. Moreover, recovering the energy from carbon captured by HRAS system would decrease the net energy consumption of the system

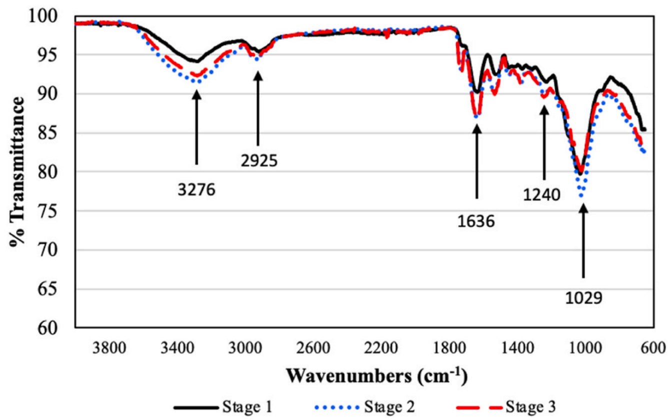

Fig. 5. FTIR spectrum of granules at each stage.

Table 3
PEPS and cEPS measurements, and pEPS/cEPS ratio at each stage.

Parameter	Unit	Stage 1 (Average ± SD)	Stage 2 (Average ± SD)	Stage 3 (Average ± SD)
pEPS	mg/g VSS	51 ± 4	56 ± 2	75 ± 6
cEPS	mg/g VSS	24 ± 2	30 ± 1	27 ± 4
pEPS/cEPS ratio	(mg pEPS/gVSS)/(mg cEPS/gVSS)	2.14 ± 0.11	1.85 ± 0.09	2.80 ± 0.24

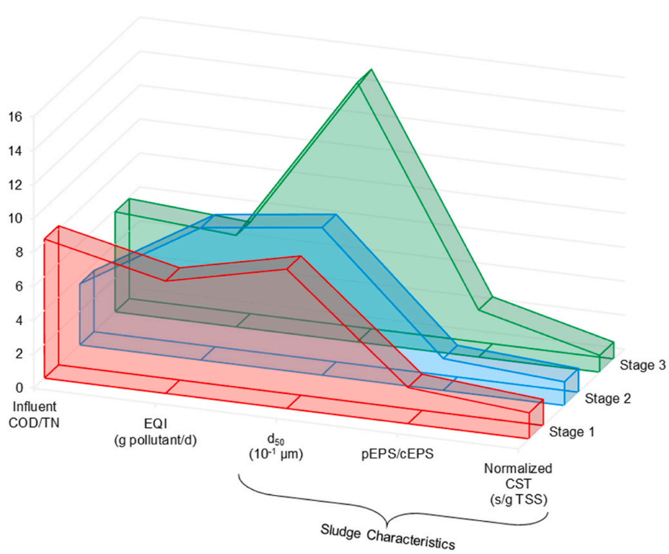

Fig. 6. Comparison of different stages in terms of EQI, d_{50}, pEPS/cEPS, and normalized CST.

that couples HRAS and AGS processes. Compared to conventional activated sludge systems, AGS process has 50% less energy consumption (Sengar et al., 2018). This makes selecting AGS process to remove nutrients after HRAS process instead of conventional activated sludge process more desirable. Regarding the coupling of HRAS process with AGS process, in order to increase denitrification performance of the system, use of volatile fatty acids (VFA) produced after fermentation of waste sludge of HRAS instead of raw wastewater in the mixing with HRAS process effluent should be investigated in the future. VFAs have shown to be an affective carbon source for granulation, phosphorus removal, and simultaneous nitrification/denitrification (Franca et al., 2018).

4. Conclusions

In this study, the performance of AGS process fed with HRAS process effluent was investigated with the aim of increasing energy recovery potential from wastewater without compromising effluent quality. AGS process was fed with raw municipal wastewater, with the effluent of HRAS process, and with the mixture of the effluent of HRAS process and raw wastewater. Although feeding the AGS process with the effluent of HRAS process lowered the influent pollution load to AGS process, low COD/TN ratio in the influent limited denitrification and caused high nitrate concentration in the effluent. The mixture of the effluent of HRAS process and raw wastewater as substrate for the AGS process increased denitrification efficiency of the system, since sufficient carbon was provided for denitrifiers with increased COD/TN ratio. The granular sludge in this system had the largest diameter, which likely also contributed to extra simultaneous nitrification/denitrification in the system. This study showed that feeding the AGS system with a mixture of HRAS process effluent and raw wastewater may be a suitable option for the treatment of municipal wastewater, while maximizing energy recovery.

CRediT author statement

Sadiye Kosar: Conceptualization, Methodology, Investigation, Writing - Review & Editing. **Onur Isik**: Conceptualization, Investigation, Validation, Writing - Original Draft, Writing - Review & Editing. **Busra Cicekalan**: Investigation. **Hazal Gulhan**: Conceptualization, Methodology, Writing - Original Draft, Writing - Review & Editing. **Seyma Cingoz**: Project administration, Funding acquisition. **Mustafa Yoruk**: Project administration, Funding acquisition. **Hale Ozgun**: Conceptualization, Supervision, Validation, Writing - Original Draft, Writing - Review & Editing, Project administration. **Ismail Koyuncu**: Resources, Writing - Review & Editing. **Mark C.M. van Loosdrecht**: Writing - Review & Editing. **Mustafa Evren Ersahin**: Conceptualization, Supervision, Validation, Writing - Original Draft, Writing - Review & Editing, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors are unable or have chosen not to specify which data has been used.

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