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Simultaneous nutrient and organic matter removal from wastewater by aerobic granular sludge process

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Abstract. Aerobic granular sludge (AGS) technology offers several benefits, such as simultaneous removal of nutrients and organic matter from wastewater, stronger granule structure, excellent settleability, and high resistance to toxicity. However, the formation of granules can take a long time and needs to be dense and stable.
In this study, the formation of aerobic granules in sequencing batch reactors (SBRs) using a granular activated carbon (GAC) and aluminium sulphate coagulant were evaluated for the simultaneous removal of nutrient (phosphorus (TP) and nitrogen (TN)) and organic matter (chemical oxygen demand (COD)) from wastewater. The reactors were continuously operated for 107 days and were fed with synthetic media and real domestic sewage. However, adaptation process with the synthetic wastewater led to relatively slow granulation process (sedimentation rate of sludge flocks was 3 m h^{-1}). During the experiments, there was no visible formation of granules in SBRs based on the analysis of the sludge samples, only the formation of aggregate structures similar to flocks. However, the results showed that total phosphorus (TP) removal efficiency was over 90% in SBR operated with aluminium sulphate. However, COD and total nitrogen (TN) removals were higher in GAC SBR, 75% and 10%, respectively. Thus, even if granules are not developed yet, the system is working efficiently. The results of this study could be useful in the development of AGS technology for full-scale wastewater treatment plant.

Key words: aerobic granular sludge, nutrient removal, organic matter, sequencing batch reactor, wastewater treatment.

INTRODUCTION

Phosphorus and nitrogen are essential nutrients that have been identified as the primary causes of excessive plant, algae, and certain bacteria growth, including cyanobacteria, in surface water bodies, resulting in eutrophication (Gorham et al., 2017; Bhagowati & Ahamad, 2018). The release of wastewater treatment plant effluents into surface waters contributes significantly to the levels of phosphorus loading in surface water bodies (Comber et al., 2013). Therefore, the efficient removal of nutrient from wastewaters are crucial. And the implementation of cost-efficient and advanced technologies is critically important to improve wastewater treatment process sustainability.

Aerobic granular sludge (AGS) is a relatively new biological wastewater treatment technology that has been investigated as a promising alternative to conventional activated sludge (CAS) process for biological wastewater treatment (De Kreuk, 2006; Pronk et al., 2015). The main advantage of the AGS technology is simultaneous removal of nutrient and organic matter from wastewater due to highly diverse microbial community in granule structure (Castellanos et al., 2021). Additional benefits are stronger granule structure, excellent settleability and high resistance to toxicity (Adav et al., 2008; Li et al., 2014). Due to these advantages, AGS technology has a great potential to be one of the most prospective biological wastewater treatment approaches in future (Zhang et al., 2016). However, the formation of granules can take a long time (Liu & Tay, 2004; Tao et al., 2017) and it is crucial to form dense stable granules. A variety of technologies may be used to enhance the formation of granules including use of coagulants (Cheng et al., 2014; Liu et al., 2014), carrier materials such as granular activated carbon (Tao et al., 2017) or adding of metal ions (Liu & Tay, 2004).

AGS process mainly has been applied in sequencing batch reactor (SBR) systems. The operation of an SBR is based on fill-and-draw principles, which typically consists of four steps-fill, aeration, settling and drawing at the same reactor. In current study, the AquaNereda® AGS technology (2017) batch cycle structure was used; where four steps of a typical SBR cycle are simplified into three steps: 1) simultaneous fill and draw operation; 2) aeration; 3) settling.

In this study, the granular activated carbon (GAC) was used as the carrier media for microbiological aggregation to initiate rapid granule formation in the SBR (Li et al., 2011). Also, the presence of Al^{3+} reduces the granulation time by approximate one month (Liu et al., 2019). As well, aluminium sulphate is widely used as an inorganic coagulant in wastewater treatment. For that reason, aluminium sulphate also was used in our study.

Current research was intended to evaluate the formation of aerobic granules in three identical SBRs: GAC SBR, $Al_2(SO_4)_3$ SBR and biological SBR (without the use of chemical precipitation) for the simultaneous removal of total phosphorus (TP), total nitrogen (TN) and organic matter (chemical oxygen demand (COD)). Therefore, the aims of the experimental study were to develop simple and effective techniques, which could accelerate the rapid granule formation and nutrient removal from wastewater. The results of this study could provide the useful information in the development of AGS technology for full-scale wastewater treatment plant.

MATERIALS AND METHODS

SBRs experimental set-up

Three identical open-type, cylindrical columns (6 cm in diameter and 50 cm in height) with a working volume of 1.55 L each were used in this study. The schematic diagram of SBR operation is illustrated in Fig. 1.

Influent was fed into the reactors from the bottom of the column at a flow rate of 50 mL min^{-1} by a peristaltic pump (MasterFlex L/S, model 77202-50, Cole-Parmer Instrument Co.). Air was added through a porous stone air diffuser (Marina 50, Hagen, China) placed at the bottom of each reactor.

The SBRs were operated in successive cycles of 3 h each. One cycle consisted of 2 min feeding and simultaneous discharge, 170 min aeration, 8 min settling. The operation of the reactors was controlled automatically by a digital process controller

(Controller, Adrona, Latvia). The effluent was at the height of 47 cm from the bottom of the reactors. pH was monitored (Multi 340i SET B, WTW, Germany), but not controlled. The SBRs were operated at room temperature 20 ± 2 °C.

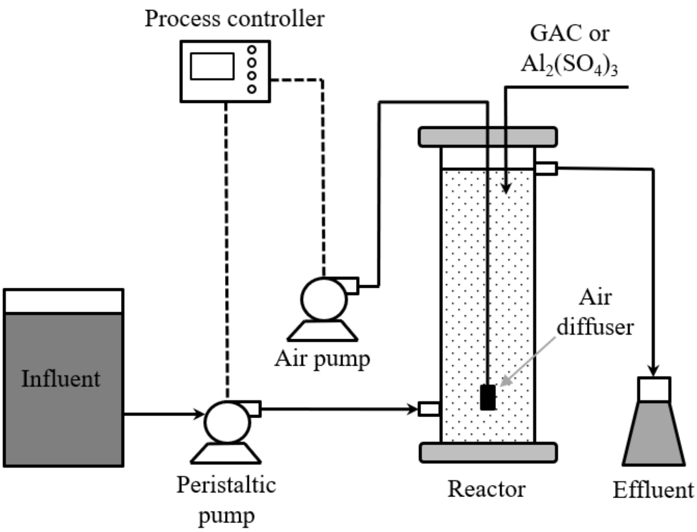

Figure 1. The schematic diagram of experimental set-up.

The nutrient (TP and TN) and organic matter (COD) removal efficiency (%) was calculated as Eq. (1) below:

$$Removal\ efficiency\ (\%) = \frac{C_0 - C_f}{C_0} \times 100\% \tag{1}$$

where C_0 – is the TP, TN, and COD influent concentrations in mg L^{-1}, and C_f – the TP, TN, and COD effluent concentrations in mg L^{-1}.

Wastewater characteristics

All three SBRs reactors were directly inoculated with 200 mL of activated sludge collected from a local the municipal wastewater treatment plant 'Daugavgriva' (Riga, Latvia). The reactors were fed with synthetic wastewater of the following composition: Media A: NaAc 63 mM, $MgSO_4$ $7H_2O$ 3.6 mM, KCl 4.7 mM; Media B: NH_4Cl 35.4 mM, K_2HPO_4 4.2 mM, KH_2PO_4 2.1 mM and 10 mL L^{-1} trace element solution. The trace element solution contained per litre: 55 g of Na_2-EDTA, 22 g of $ZnSO_4{\cdot}7H_2O$, 5.54 g of $CaCl_2{\cdot}2H_2O$, 5.06 g of $MnCl_2{\cdot}4H_2O$, 5 g of $FeSO_4{\cdot}7H2O$, 1.1 g of $(NH_4)Mo_7O_{24}{\cdot}4H_2O$, 1.57 g of $CuSO_4{\cdot}5H_2O$, 1.61 g of $CoCl_2{\cdot}6H_2O$, adjust pH to 6.0 with KOH according to Vishniac & Santer (1957).

SBRs operation strategies

Three different operation strategies were tested to study the granulation process nutrient and organic matter removal from wastewaters by AGS technology. Biological SBR was used as a 'control' - biological process without the use of additions; $Al_2(SO_4)_3$ SBR was operated with $Al_2(SO_4)_3$ coagulant; and GAC SBR was run with granular activated carbon. The reactors were continuously operated for 107 days, and the operation time was divided in three stages, which represented different synthetic and real wastewater concentrations. Detailed information about the SBR stages and feed is shown in Table 1.

Table 1. Operational Stages according to changes in influent ranges of synthetic medium and real wastewater ratio. Composition of influent

Stage	Time, days	Proportion in influent, %		Total P, mg L^{-1}	Total N, mg L^{-1}	COD, mg L^{-1}
		Synthetic wastewater	Real wastewater			
I	0–14	100	0	124 ± 10	39 ± 1	459 ± 10
II	15–35	95	5	118 ± 14	43 ± 4	378 ± 13
III	43–107	90	10	57 ± 11	30 ± 4	277 ± 20

Aerobic sludge granulation is rather difficult to achieve for the treatment of a real municipal wastewater. For this reason, adaptation (constant reduction of concentration of media and increase of concentration of real wastewater) of sludge to wastewater was performed from the beginning of the experiment.

In the first 14 days, stage I, the reactors were started up with the synthetic wastewater. During the next two operation stages, the dosage of synthetic wastewater was reduced by 10%, 5% reduction in each of the operation stage. At the same time, the dosage of real wastewater was increased.

Selection of coagulant

A series of jar tests were carried out to determine which coagulant was most suitable for pre-treatment of the wastewater used in this study. Jar tests were performed using a synthetic wastewater consisting of (by volume) 20% activated sludge and 80% synthetic wastewater. Four coagulants were evaluated: magnesium chloride ($MgCl_2$), aluminium sulphate ($Al_2(SO_4)_3$), calcium chloride ($CaCl_2$) and ferric chloride ($FeCl_3$). The optimal coagulant dose was determined by adding different coagulant concentrations ranged from 5 to 150 mg L^{-1}. pH (Multi 340i SET B, WTW, Germany) and zeta potential (Zetasizer Nano ZS90, Malvern) were determined to find an optimal coagulant dosage for an effective coagulation and sedimentation. The most effective coagulation process is achieved when zeta potential is about +/- 0.5 mV (DeWolfe et al., 2003; Lopez-Maldonado et al., 2014).

Bio-carrier

Granular activated carbon (GAC) was selected as the support medium for faster granulation. 2 g L^{-1} of GAC (Chemviron, Norrlandskol, 0.4–1.4 mm) was used for experiments.

Analytical methods

The influent and effluent samples were collected once a week and concentrations of total chemical oxygen demand (COD), total nitrogen (TN), total phosphorus (TP) were measured using portable data logging colorimeter (DR/890, HACH). The pH value, electrical conductivity (EC), RedOx potential measurements were performed using Multi 340i SET B equipment (WTW, Germany). Zeta potential was measured using Zetasizer Nano ZS90 (Malvern, UK). The morphology of the sludge samples was visualised by light microscopy (Leica 6000B, Germany) equipped with Image-Pro Premier software.

The sludge samples were collected from the reactors, homogenized by gentle shaking, and applied to clean glass slides.

RESULTS AND DISCUSSION

Preliminary evaluation of coagulants

A preliminary evaluation of the coagulants was carried out in jar tests in order to found out the optimum coagulant type and coagulant dosage. The relation of different coagulants on the Zeta potential is shown in Fig. 2.

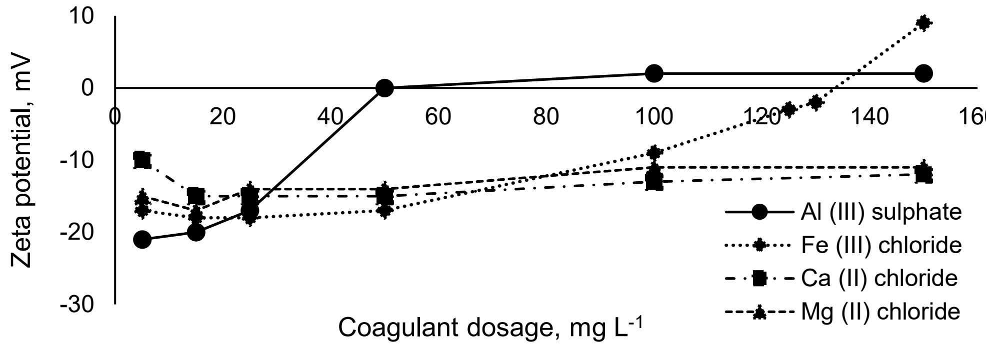

Figure 2. The effect of coagulant type and coagulant dosage on zeta potential. Jar tests performed using (●) $Al_2(SO_4)_3$, (♦) $FeCl_3$, (■) $CaCl_2$ and (▲) $MgCl_2$ coagulants in concentration range from 5 to 150 mg L^{-1}.

The results of the evaluation of different coagulant types indicated that the point of zero charge was observed only for $Al_2(SO_4)_3$ at 50 mg L^{-1} Al^{3+}; and close to the zero value (-2 mV) for $FeCl_3$ coagulant at 130 mg L^{-1} Fe^{3+} (Fig. 2). $MgCl_2$ and $CaCl_2$ coagulants did not reach the point of zero charge during the jar tests at coagulant doses ranged from 5 to 150 mg L^{-1}. Therefore, $MgCl_2$ and $CaCl_2$ coagulants were omitted from sludge settling experiments.

Sludge settling procedure

The sludge settling assessment was performed at optimal dose of $Al_2(SO_4)_3$ and $FeCl_3$ coagulants (50 mg L^{-1} and 130 mg L^{-1}, respectively) (Fig. 2).

Sludge settling experiments were performed using synthetic wastewater consisting of (by volume) 20% activated sludge in a 1 L graduated glass cylinder. The volume of activated sludge was measured visually at 0, 1, 5, 10, 20, 25 and 30 minutes. Fig. 3 illustrates sludge settling curves for control sample (without the use of chemical precipitation) and samples with coagulants: for the aluminium sulphate at 50 mg L^{-1} Al^{3+} and ferric chloride at 130 mg L^{-1} Fe^{3+}.

The results (Fig. 3) showed that 130 mg L^{-1} ferric chloride coagulant has better sludge settling rate than 50 mg L^{-1} aluminium sulphate. At the same time, the sludge settling rates of the control sample (without coagulant) and ferric chloride were similar during the sludge settling experiment. Although, ferric chloride showed better results on sludge settling rate, aluminium sulphate was used in sequencing batch reactor due to availability and financial reasons.

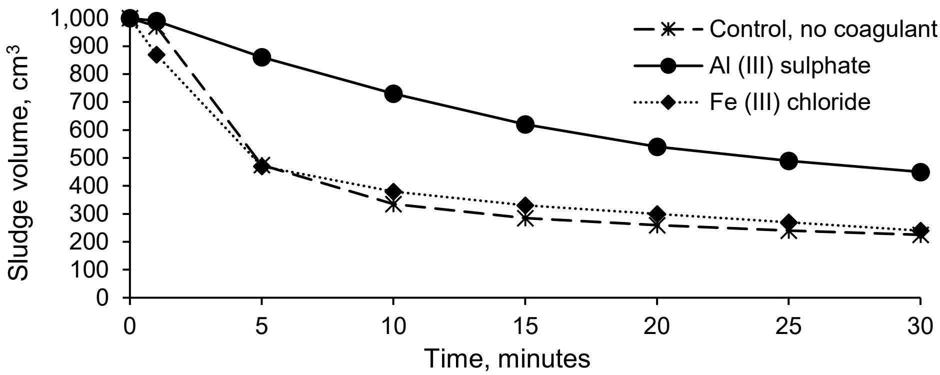

Figure 3. Sludge settling curves. Tests performed using synthetic wastewater consisting of 20% activated sludge with aluminium sulphate (●, 50 mg L^{-1} Al^{3+}), ferric chloride (♦, 130 mg L^{-1} Fe^{3+}), and control (⁎ , no coagulant).

Sludge characterization with microscopy

Aerobic granules are described as approximately spherical aggregates of a size larger than 600 µm and consisting of mostly bacteria and extracellular polymeric substances. The average density has been reported varying from 30 to 150 g L^{-1} biomass (Lemaire et al., 2008).

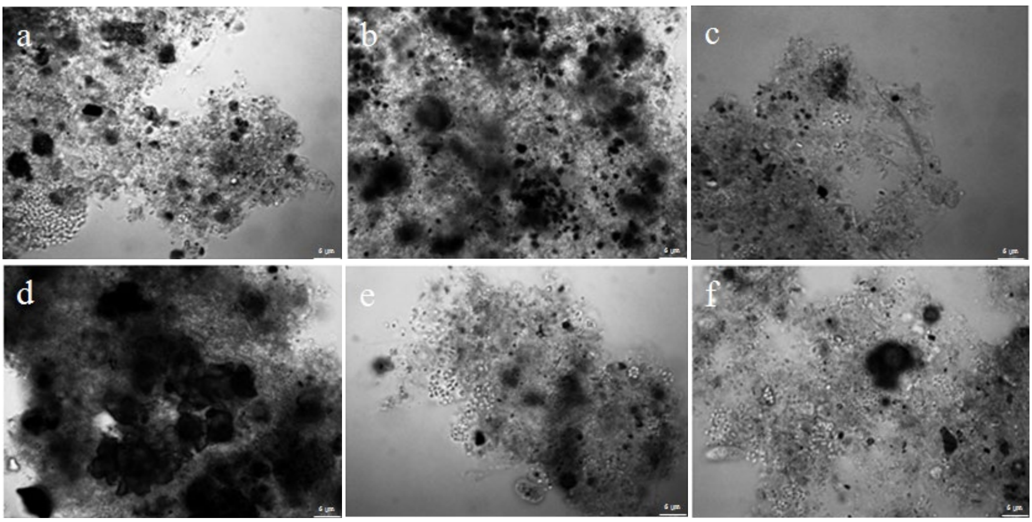

Figure 4. Microscopy images of the sludge during operation in the SBRs: (a) 14 days, (b) 107 days in control SBR; (c) 14 days, (d) 107 days in $Al_2(SO_4)_3$ SBR and (e) 14 days, (f) 107 days in GAC SBR.

As can be seen in Fig. 4, the initial seeding sludge (day 0) had fluffy and irregular structure (Fig. 4, a; 4, c; 4, e). After 107 days of the experiments, the seeding sludge with a fluffy structure gradually disappeared and was replaced by small flock type aggregates (Fig. 4, b; 4, d; 4, f). This could be explained by the fact that the selective discharge of small sludge flocks from the SBRs have not been applied during the experiments. Similar results were reported by Yang et al. (2004) and Li & Li (2009), who observed that small and loose flocs can obtain substrates more easily from the suspension, which allows them to grow faster. As a result, without selective sludge discharge, there is less substrates available for uptake by small flocks, which means that it is not possible to developed stabile and dense granules.

Nevertheless, all samples contained high variety of microorganisms - various shaped bacteria, protozoa and uncharacteristic shapes attributable to non-cellular material. As reported Weber et al. (2007), one of important factors in facilitating flock formation is the presence of ciliates and fungi that serve as the main substratum in the formation of bacterial biofilms and act as a backbone for the granules. Thus, the microbial diversity and community structure could play an important role in aerobic granular sludge formation.

Nutrient (TP and TN) and organic matter (COD) removal in SBRs

The operational results of three monitoring reactors of TP and TN removal efficiencies are illustrated in Fig. 5 and Fig. 6, respectively. $Al_2(SO_4)_3$

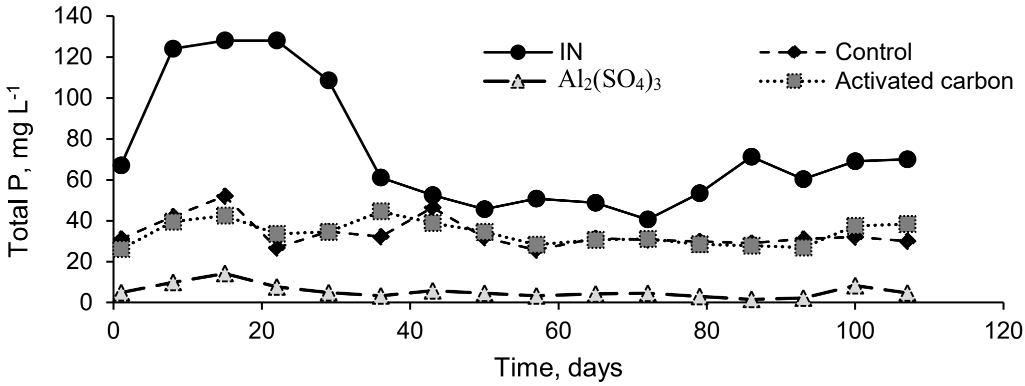

Figure 5. Total P removal efficiencies for control SBR, ($Al_2(SO_4)_3$) SBR and GAC SBR during 107 days.

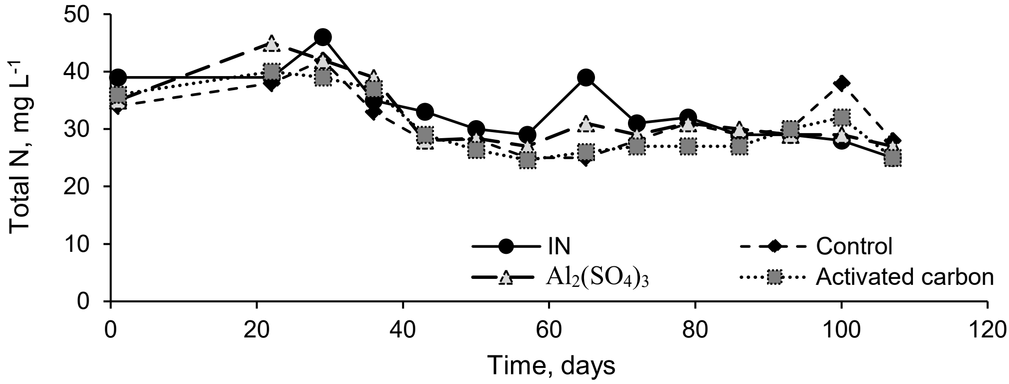

Figure 6. Total N removal efficiencies for control SBR, ($Al_2(SO_4)_3$) SBR and GAC SBR during 107 days.

The results showed (Fig. 5; Table 2) that the highest phosphorus removal rate (90%) was in $Al_2(SO_4)_3$ SBR. Similar results of TP removal efficiency were obtained by Liu et al. (2019) using aluminium based coagulant. However, nitrogen removal rates were lower than 10%in all three reactors during all operation period (Fig. 6; Table 2). Previous studies have indicated that biodegradable carbon source availability is required to maintain simultaneous phosphorus and nitrogen removal in the SBRs (Kong et al., 2014; Lashkarizadeh et al., 2015).

Table 2. Nutrient and organic matter removal efficiencies in the SBRs in different Stages of operation

Parameter	Stage I	Stage II	Stage III	Average (107 days)
TP removal (%)				
Control SBR	62	71	47	60 ± 12
$Al_2(SO_4)_3$ SBR	92	93	93	93 ± 1
GAC SBR	66	70	44	60 ± 14
TN removal (%)				
Control SBR	12	8	4	8 ± 4
$Al_2(SO_4)_3$ SBR	9	0	4	4 ± 3
GAC SBR	6	8	10	8 ± 2
COD removal (%)				
Control SBR	49	70	69	63 ±12
$Al_2(SO_4)_3$ SBR	54	71	71	65 ± 10
GAC SBR	66	73	75	71 ± 5

During the stage I, the reactors were fed with synthetic wastewater, influent COD concentration was about 460 mg L^{-1} (Table 1). The COD removal rates for control SBR, $Al_2(SO_4)_3$ SBR and GAC SBR were 49%, 54% and 66%, respectively (Fig. 7; Table 2).

In the stage II, the dosage of synthetic wastewater was reduced by 5% and influent COD concentration decreased to 380 mg L^{-1}, average TP removal efficiencies for control SBR, $Al_2(SO_4)_3$ SBR and GAC SBR were 71%, 92% and 70%, respectively, which were the highest phosphorus removal efficiencies in the three SBRs for 107 days. The nitrogen removal efficiencies for control SBR and GAC SBR were similar - 8%. However, no TN removal observed in $Al_2(SO_4)_3$ SBR. The COD removal efficiencies of 70%, 71% and 73% were achieved by SBR, $Al_2(SO_4)_3$ SBR and GAC SBR, respectively.

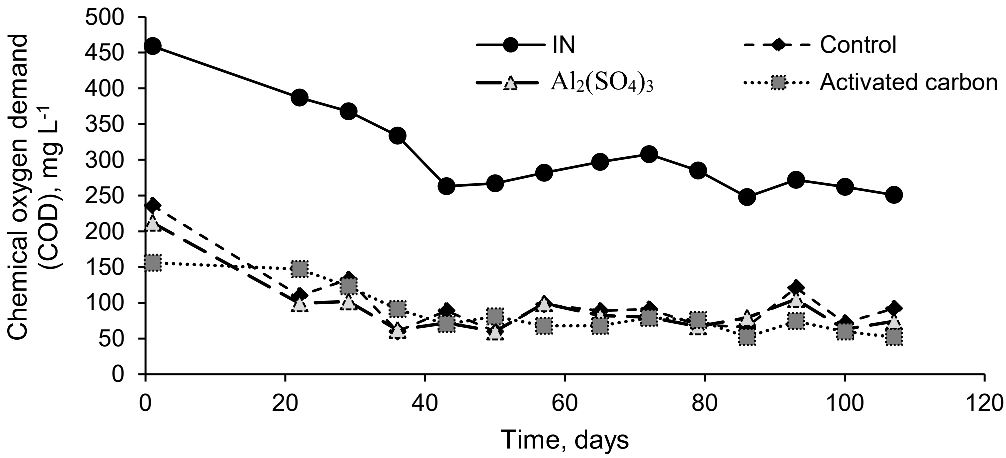

Figure 7. COD removal efficiencies for control SBR, ($Al_2(SO_4)_3$) SBR and GAC SBR during 107 days.

In the stage III, the dosage of synthetic wastewater was reduced by 10% and influent COD concentration was decreased to 280 mg L^{-1}, average TP removal efficiencies were 47%, 93% and 44% for control SBR, $Al_2(SO_4)_3$ SBR and GAC SBR, respectively. At this stage, the lowest phosphorus removal efficiencies (less than 50%) were observed for control SBR and GAC SBR after the change in the feed. This was caused by the reduction in influent phosphorus concentration from 124 to 57 mg P L^{-1}. It should be noted that the effluent concentration of TP has not decreased after the change in the feed.

During 107 days, the average TN removal efficiency was 8 ± 4% for control SBR, 4 ± 3% for $Al_2(SO_4)_3$ SBR and 8 ± 2% for GAC SBR. Despite the differences in three reactors, removal efficiency of COD was similar. The average COD removal efficiency was 63 ± 12% for control SBR, 65 ± 10% for $Al_2(SO_4)_3$ SBR and 71% for GAC SBR. The average TP removal efficiency was 60 ± 12% for control SBR, 93 ± 1% for $Al_2(SO_4)_3$ SBR and 60 ± 14% for GAC SBR. The results showed that control SBR and GAC SBR had the same TP removal efficiency. Therefore, TP removal efficiency (more than 50%) can be achieved in biological wastewater treatment systems without the adding of GAC in SBR. These values are similar to the reported previously (Denisova et al., 2020), where phosphorus removal efficiency was 88% using pilot-scale SBRs even without the development of stable aerobic activated granules.

CONCLUSIONS

In the present study, Al^{3+} and GAC were used to improve the granule formation in SBRs for simultaneous nutrient and organic matter removal from wastewater. The highest removal efficiency of TP was over 90% in SBR operated with aluminium sulphate. However, COD and TN removals were higher in GAC SBR, 75% and 10%, respectively. Although full granulation was not achieved. Moreover, the ease of maintenance and operation are the advantages of using SBRs in full-scale wastewater treatment plants. However, further research is needed to obtain more stable and high-efficiency removal performance of simultaneous nutrient and organic matter from wastewater by aerobic granular sludge process.

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