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Cultivation of aerobic granules through synthetic petroleum wastewater treatment in a cyclic aerobic granular reactor

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ABSTRACT

The aim of this study was to investigate the new structure of SBR (placement of baffles in different way) and its ability to treat petroleum wastewater, compared to a conventional system, as well as investigate the effects of pollutant concentration on the granules' performance at ambient temperature (24 ± 1ºC). Two SBR were tested. R_1 had baffles including three risers and three down-comers, and R_2 had one riser and one down-comer. After an adaptation period of about 59 d, granules with a mean diameter of 6 mm and 5 mm, medium density of 1.0132 and 0.87720 gr/ml, and settling velocity of 2.51 and 2.13 cm/s were observed in R_1 and R_2, respectively. Stepwise increasing the OLR in R_1 and R_2, affected size, density, settling velocity, and the granules' stability, by increasing them and gradually decreasing the COD removal efficiency. Maximum COD removal in R_1 and R_2 was achieved in OLR equal to 0.9 kg/m^{3}d (95.4%–91.4%), and minimum COD removal was achieved in OLR equal to 2.4 kg/m^{3}d (85.5–71.1%). Corresponding amounts for maximum removal of total petroleum hydrocarbon (TPH) were 94.2% and 90.8% (TPH_{in} equal to 104.2 mg/l) and minimum amounts (TPH_{in} equal to 157.9 mg/L) were equal to 82.1% and 70.4%, respectively. Appropriate placement of baffles in R_1 caused faster formation of granules, improved their physical properties and increased removal efficiency compared to R_2. By increasing in OLR and consequently, increasing in granule size and according to cell death, an anaerobic core generated in the center of the granules. Instability of anaerobic core as well as reduction of the granules' ability to tolerate oil and toxic pollutants, gradually decreased removal efficiency, integrity coefficient and increased SVI.

Keywords: COD; Aerobic granules; Organic loading; petroleum wastewater; Sequencing batch reactor (SBR)

1. Introduction

A large volume of water is used for refining crude oil in petroleum refineries, producing a great amount of wastewater. The composition of these wastewaters differs and depends on the crude oil and purification processes [1]. Petroleum compounds are classified into two main groups: (1) the hydrocarbons, which consist of paraffin, naphthene, and aromatics, and (2) other compounds, such as oxygen, nitrogen, sulfur, and rare metal atoms such as nickel and vanadium [2].

The high molecular weight of polynuclear aromatic hydrocarbons (PAHs) is known to cause mutations and cancer, while lightweight PAHs have acute toxicity [3]. Since 1979, the United States Environment Protection Agency (EPA) has considered PAHs as priority pollutants [4]. Therefore, it is necessary to treat these compounds before releasing them into the environment.

Conventional treatment methods of petroleum wastewater include different physical methods such as adsorption [5], physical separation [6], membrane filtration [7], flotation [8], centrifuge [9], freezing [10], chemical methods such as coagulation and flocculation [11], electrochemical

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processes [12], advanced oxidation [6], biological methods such as activated sludge systems [13], UASB [14], SBR [15], and MBBR [16].

Granular sludge is one of the most useful technologies for wastewater treatment [17]. In this technology, aerobic particles, which formed by self-aggregation processes, cause appropriate solid-liquid separation [18]. A workshop held in Germany in 2004 defined aerobic granules as microbial origin aggregates with a high settling velocity as compared to activated sludge flocs, which have tremendous stability against coagulation under low hydrodynamic shear. In the last decade, the results of several studies indicate that aerobic granular sludge technology is an evolution in the biological treatment of wastewater [19]. Characteristics of aerobic granules can be described by morphology, settle ability, density,physical strength, cell surface hydrophobicity, and storage stability [20]. Aerobic granule reactors have attracted direct attention because of their excellent sludge settleability, shock loading resistance ability [21], long sludge retention time, and no sludge bulking [22]. Factors affecting granulation include seed sludge, substrate composition, organic loading rate, feeding strategy, reactor operation, settling time, exchange ratio, aeration intensity, and environmental factors such as temperature and pH [23].

Successful performance of this technology have been reported in several studies. Derlon et al. used SBAR for treating municipal wastewater. They have reported high soluble COD (SCOD) removal efficiencies (>80%) after 4 h operation. Average input SCOD concentration was between 127 ± 55 mg/L [17]. Ni et al. treated low-strength municipal wastewater (<200 mg L^{-1}) of COD with use of SBR technology. They have reported that average COD removal efficiency was approximately 90% after the formation of aerobic granules [19]. Pajoumshariati et al. assessed MSBR[1] for petroleum refinery wastewater treatment. After 95 d operation, average COD and TPH removal efficiencies were 80 and 93.4%, respectively. Input COD was in the range of 43.4–335.3 mg/l [24].

According to successful granulation in conventional SBR (with riser and down-comer), the objective of the present study was investigation of the SBR system's new design (placement of the baffles in different way) in treating oily wastewater (contain petroleum contaminants). The main focus was on the challenges associated with the biogranules, including (i) effects of baffles compared to conventional systems on the granulation time and characteristics of biogranules; (ii) determination of the maximum tolerable concentration of COD and TPH in fed-batch condition; (iii) investigation of granules' ability to withstand organic shock loading. Further, the granules were investigated for physical characteristics such as settling ability, density, physical strength in the presence of toxic compounds.

2. Materials and methods

2.1. Reactor setup and operation

In this study, two parallel Plexiglas reactors, R_1 and R_2, were operated under aerobic conditions.

Table 1 describes the characteristics and operation phase for R_1 and R_2. R_1 had baffles including three risers and three down-comers, and R_2 had one riser and one down-comer. The unique characteristic of R_1 was its liquid flow rotation in the down-comer, which connected the top and bottom of the reactors. Due to the high flow rotation, effective mixing and suitable oxygen transfer occurred without use of a mechanical stirrer [25]. Fig. 1 shows the schematic of the operated pilots. The systems were run on a 6-h cycle. Cycles were carefully controlled in timed steps by a PLC[2]. The PLC was connected to a supervisory control and data acquisition (SCADA) system. The discharge port was 28 cm above the base of the reactors, achieving a 50% volumetric exchange ratio per cycle.

2.2. Methods

Activated sludge was collected from a municipal wastewater treatment plant in Tehran, Iran, and used as seed sludge for the reactors. Initial pH, temperature, DO, MLSS and MLVSS of the seed sludge (before injection into reactors) were 7.2, 20°C, 3.4 mg/L, 3900 mg/L, and 3200 mg/L respectively.

To achieve a similar wastewater composition to that of the Tehran petroleum refinery, the combination of gasoline (C_{16}–C_{20}) and crude oil (C_8–C_{37}) with a volume ratio (v/v) of 1:2 was used. The mixture aerated for 48 h. By doing this, the light compounds exited with the water vapor and entered the gas phase and heavy hydrocarbons (heavier than C_{35}) congealed in the container. This process prevented the entrance of too heavy and too light hydrocarbons to the reactors [26]. Input COD (COD_{in}) of the synthetic wastewater for each cycle, calculated and adjusted by the C/N/P ratios (on a molar basis) at 100:5:1 and then fed to the reactors. CH_4N_2O, KH_2PO_4, K_2HPO_4 and glucose were used as the nitrogen, phosphorus, and carbon sources, respectively. Using KH_2PO_4 and K_2HPO_4, not only supplied needed phosphorus in the reactors, but also acted as buffer against large change in the internal pH. A 1 mL·L^{-1} trace element solution contained $FeCl_3$, $MnSO_4·H_2O$, $ZnCl_2$, $CuCl_2$, $AlCl_3$, $CoCl_2·6H_2O$, $MgSO_4·7H_2O$, and $CaCl_2$ [27]. The bioreactors operated in 3 main stages included: (1) adaptation period (59 d, COD_{in} = 200 mg/L), (2) increasing the OLR (90 d, COD_{in} = 225–600 mg/L) and (3) shock induction (10 d, COD_{in} = 2400 mg/L).In the first step of adaptation (on days 1–5), injected synthetic wastewater, contained glucose with an initial COD of 200 mg/L. After complete adaptation of microorganisms, the concentration of glucose reduced by 10% (became equal to 180 mg/L) and the concentration of petroleum increased by the same percentage (became equal to 20 mg/L) to maintain the concentration of COD_{inat} 200 mg/L. This procedure was repeated to achieve a constant value in COD removal efficiency, ensuring that the microorganisms adapted to the corresponding concentration. These steps continued by increasing the concentration of petroleum to 20%, 30%, and 40% and by reducing the glucose to 80%, 70%, and 60%, so long as the 100% of the injected feed at the end of adaptation period (on days 56–59) contained 200 mg/L petroleum compounds. After achieving a steady state in the final stage of adaptation, COD increased to

[1]Membrane sequencing batch reactor

[2]Programmable logic controller

Table 1
Characteristics and operations for R_1 and R_2

Configuration	R_1	R_2	Operation phase for R_1 and R_2	Time (min)
Average height (cm)	60.5	60.5	Feeding	3
Length (cm)	19.3	19.3	Aeration	310
Width (cm)	4.8	4.8	Settling	25
Riser's dimension (cm)	4*4	4*10.5	Discharge	7
Down-comer's dimension (cm)	1.5*4	3*4	Idle	15
Working volume of pilots (L)	3.6	3.6	Total cycles per day	360

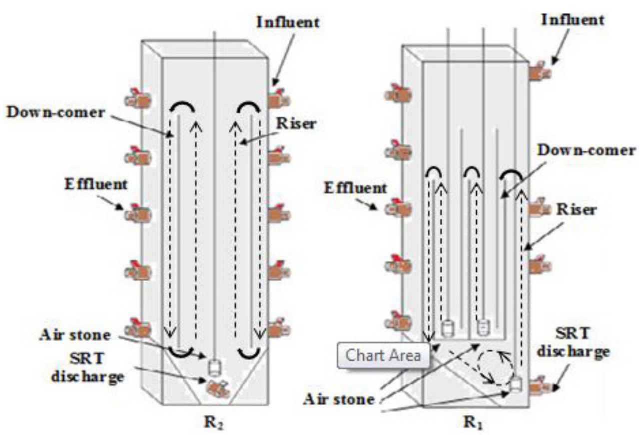

Fig. 1. Schematic drawing in profile of pilot scale sequencing batch reactors and flow direction.

225 mg/L per cycle (equal to 0.9 Kg/m^{3}d). The input COD of the reactors was then increased stepwise by 25 mg/L to reach 500 mg/L, then again by 50 mg/L up to 600 mg/L.

During the study, pH and DO were measured regularly. By using buffers (K_2HPO_4 and KH_2PO_4), pH maintained in the range of 6.8–7.2 and DO was kept in the range of 2–6 mg/l, respectively. The laboratory's ambient temperature was in the range of 24 ± 1°C. An aquarium air pump (RS-8801) supplied needed oxygen in the reactors.

2.3. Analytical methods

Soluble COD was determined by the closed reflux, colorimetric method-5220D using standard methods [28]. A COD reactor (Hach-DR4000) was used after filtering the samples through 0.45 µm cellulose filters (Millipore). TPH was determined using (TOG/TPH Analyzer-Infracal). The formation and alternation of aerobic granules were frequently monitored using a binocular optical microscope (Nikon-YS100). The granules' physical properties including settling velocity and integrity coefficient (%), which indirectly indicates the physical stability and compactness of the granules, were measured using Orbital shaker-TM52, according to the procedure described by Rosman et al. [29]. Storage stability was determined following the procedure detailed by Tay et al. [30], and density was performed according to Taheri et al. [31]. SVI and MLSS were determined at the end of the aeration phase, and the image analysis method was used for measuring the granules' size and distribution. Finally, a GC/MS analysis (Agilent-7890A/5975C) was performed for identifying intermediate compounds, and transmission electron microscopy (Zeiss- EM10C-80 KV) was conducted according to Tang et al. [32] for observing the granules' micro structure and microbial compositions. All tests were performed at ambient temperature (24 ± 1ºC), and each experiment was performed three times. The error rate was equal to 4.2%.

3. Results and discussion

3.1. Reactor operation

The removal efficiency of COD in R_1 and R_2 on day 59 reached 95.4% and 92.9%, respectively. Input OLR, COD and TPH in this stage was equal to 0.8 Kg/m^{3}d, 200 and 53.6 mg/L, respectively. In this situation, the TPH removal efficiency of R_1 and R_2 were 95.5% and 93.2%, respectively. After complete adaptation and granulation (during 59 d), the reactors operated for 90 d with the initial and final OLR equal to 0.9 and 2.4 kg/m^{3}d. To achieve a better comparison throughout the entire study, MLSS was kept in the range of 3500 mg/L. TPH tests were performed when the COD removal efficiency in each stage, reached a constant value (steady state). The overall performance of the reactors can be seen in Fig. 2. By increasing OLR in each stage, an average of 5.2 d were needed to reach steady state. When input COD (on day 84) was equal to 325 mg/L, the COD removal efficiency reached the steady state after 4 d, and the loading rate immediately increased to 350 mg/L. This caused disintegration and reduction of granules number. Consequently, the removal efficiency of COD in R_1 and R_2 reduced to 77 and 74.3%. The related reduction in TPH removal, were equal to74.1 and 72.4%, respectively. In order to improve the condition, the loading rate in the reactors decreased to 325 mg/l. In doing so, after 5 d removal efficiency of COD in R_1 and R_2increased to 91 and 82.8%, and TPH removal reached the constant value of 88.8 and 85.1%, respectively. Although, the systems experienced disintegration of large sized granules rather than the small one, removal efficiency increased quickly. This implies that smaller size granules, benefits the aerobic treatment in maintaining good performance stability with high treatment efficiency. After an interruption in this stage, the input organic load increased to 350 mg/L (1.4 kg/m^{3}d).The maximum COD and TPH removal were achieved when OLR was equaled to 0.9 kg/m^{3}d. The maximum removal values for COD in R_1 and R_2were 95.4 and 91.4% and maximum removal of TPH were 94.2%

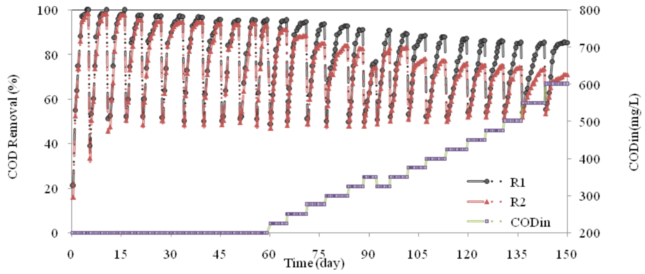

Fig. 2. COD removal efficiency in reactors (OLR_{in} = 0.8–2.4 Kg/m³d).

and 90.8%, respectively. Moreover, the minimum removal was achieved when OLR equaled 2.4 kg/m³d. In the same way, the minimum removal values for COD in R_1 and R_2 were 85.5 and 71.1% and minimum removal of TPH were 82.1% and 70.4%, respectively. The figures show that the existence of baffles in R_1 improved the system's performance and increased removal efficiency. By increasing in hydrodynamic shear force (due to the existence of baffles), sludge moved along a greater path and rotated more, and this caused formation of denser granules and increased the removal efficiency. As shown in Figs. 2 and 3, removal efficiency decreased as the pollutants' concentration increased, which could be due to reduction in the sludge's ability to tolerate the toxic pollutant. The results show that granular SBR technology can be as efficient as prospective technology for treating hard biodegradable components.

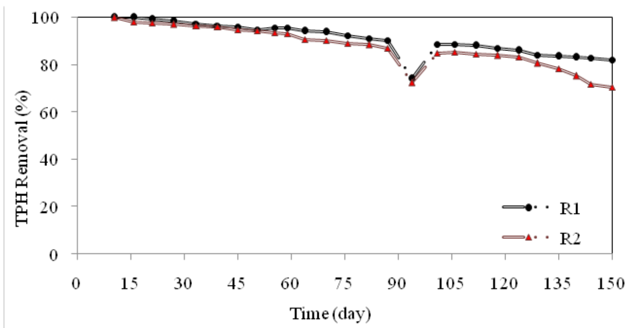

Fig. 3. TPH removal efficiency in reactors (TPH_{in} = 0–157.9 Kg/m³d).

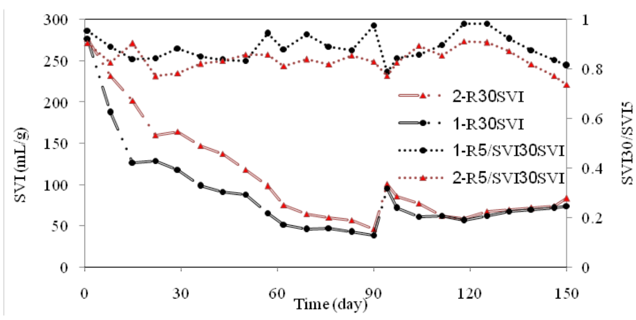

Fig. 4. Variation of SVI_{30} and SVI_{30}/SVI_5 during operation.

3.2. Sludge volume index

Sludge volume index (SVI) was used as an index for the sludge granulation value. Values of SVI_{30} for mature granules have been reported in the range of 20–60 ml/g [33]. Moreover, reduction in SVI represents formation of granules. Kusmierczak et al. [34] reported less difference between SVI_5 (or SVI_{10}) and SVI_{30} in granular sludge compared with flocculated sludge, and also reported the stabilized SVI between 90–110 mL/g. Therefore, as Liang et al. [33] report, SVI_{30}/SVI_5 is another index for granulation rate. In this study, the tests on primary sludge indicate high values for SVI_{30} (277 ml/g) and SVI_5 (297 ml/g). As can be seen in Fig. 4, the overall profiles of SVI_{30} show a downward trend until day 90, and reached the lowest amount of 38 and 45 mL/g, respectively. This result confirmed that the biogranules had better settle ability than the flocs. During this period, SVI_{30}/SVI_5 was in the range of 0.83–0.97 and 0.82–0.95, respectively. Results indicate that the settling performance of the cultivated granules improved by increasing the OLR. On day 94, the rapid increase in OLR from 1.1 to 1.2 Kg/m³d (increasing COD from 325 to 350 R_1 and R_2 mg/L), increased the SVI_{30} in R_1 from 38 to 95 mL/g and also increased in R_2 from 45 to 101 mL/g. The reason for this increase was the granules' intolerance to the input OLR, and consequently their disintegration. By increasing the OLR from 1.7 to 2.4 Kg/m³d, the overall profiles of SVI_{30} kept rising from day 125 until the end of the study and increased gradually from 62 and 67 in R_1 and R_2 to 74 and 83 mL/g, respectively. SVI_{30}/SVI_5 decreased from 0.98 and 0.94 to 0.81 and 0.77 due to the deterioration in the granules' ability to with stand oil and toxic compounds, and finally their disintegration. Because of the high granulation rate in R_1, it had less SVI and more SVI_{30}/SVI_5 compared to $R_{2,}$ meaning that granules in R_1 exhibited better settling properties than those in R_2.

3.3. Characteristics of granules

3.3.1. Average particle size

Primary sludge had fluffy, loose, irregular-shaped, gray coloured flocs (Fig. 5a). Small granules became visible in R_1 and $R_{2,}$ 5 and 7 d after inoculation, respectively (Figs. 5b and 5c). Cultivated granules were round in shape and different from the sludge flocs and gradually turned dark brown over time. The morphology and characteristics of the granules varied on different d. By the end of the adaptation period (on day 59), the mean diameter of the granules was about 6 mm and 5 mm in R_1 and R_2. These amounts gradually increased over time with the growth of the granules, reaching 8–10 mm and 7–9 mm in R_1 and R_2, respectively. High OLR and a strong shear force produced by aeration, caused the cultivation of larger sized granules. Figs. 5d and 5e show the granules' size on day 145.

Fig. 6 shows the granules' growth process and the changes in their diameter during the operation of the reactors. The mean diameter of the granules in R_1 was more than those in R_2 throughout the whole operation period. The difference was always in the range of 1–2 mm. The largest observed granule had a 12 mm diameter.

In this study, the changes in the granules' size were evaluated on six different days. Since the mean difference in the average size of the granules in the two reactors was

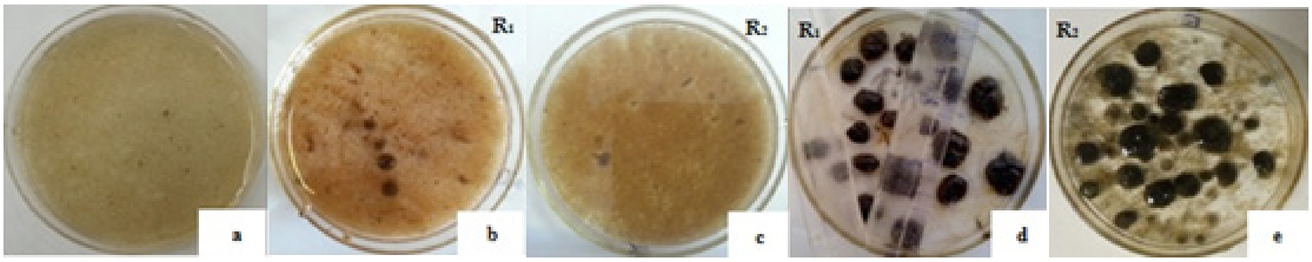

Fig. 5. Images of seed sludge (a), the first granules in R_1 on day 5 (b) and in R_2 on day 7 (c), existing granules on day 145 in R_1 (d) and in R_2 (e).

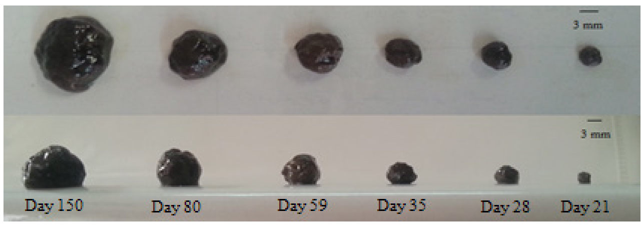

Fig. 6. Changes in granule's diameter within the period of experiments.

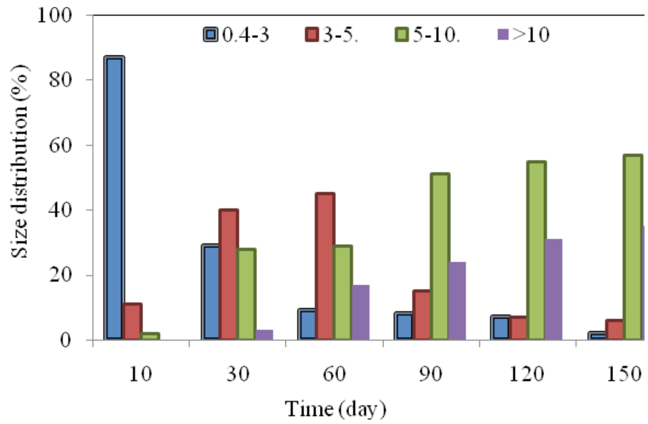

Fig. 7. Aerobic granule's size distribution in R_1 and R_2.

always between 1–2 mm, the size distribution curve for the two reactors was the same, as shown in Fig.7.

By day 10, granules with approximately 0.5–4.0 mm in diameter were dominant in the systems. This range increased to 3–5 mm on days 30 and 60. On days 90, 120, and 150, granules with an average diameter of 5–10 mm were dominant. The most significant difference among these days was in the percentage of granules with an average size of more than 10 mm, which increased gradually. Variations of granules in the range of 5–10 mm were slight on days 30 and 60, but increased sharply on day 90 when they finally measured in the beneficial range. These results have indicated that the overall profile of granules in the 0.4–3 mm range had a downward trend throughout the entire 150 d in forming larger sized aerobic granules. Generally, larger granules were more susceptible to disintegration due to their tendency to mix under high shear force and also small granules had low settling velocity. As Dahalan et al. [35] reported, the granules' high density and settling velocity are suitable indicators of successful granulation.

TEM imaging was performed to understand the details of the granules' microscopic structures. A mature granule sample was selected and investigated on d 150. As can be seen in Fig.8, various cocci, rod-shaped, filamentous cells, and alcanivorax bacteria were predominant. Kong et al. [36], also reported rod-shaped bacteria and filamentous cells, which were similar to the cultivated granules in this study.

3.3.2. Density

In environmental engineering, density is typically used to describe the structural compaction of the microbial community [34]. According to Fig. 9, when OLR is increased, the density of the granules also increased and their overall profile shows an upward trend over time. The low density of some granules were due to high empty space inside them, thereby increasing their size. The mean density of the granules in R_1 and R_2 were in the range of 0.0354–1.2225 g/mL and 0.0251–1.2008 g/ml, respectively. The difference in the density values in the two reactors was due to adequate rotation and compression in R_1 and consequently, the formation of denser granules toward R_2. The aeration rate and existence of baffles play a crucial role in the production of hydrodynamic shear force and improving the compaction of the granules.

For granules with a diameter in the range of 0.5–0.9 mm and settling velocity of 0.5–2.5 cm/s, Kuśmierczak et al. [34] reported a mean density of 1.008 and 1.004 g/ml for granules and conventional sludges, respectively. Li et al. [37], reported density in the range of 1.082 ± 0.048 g/ml for granules with a diameter and settling velocity in the range of 2.47 ± 0.97 mm and 1.69 ± 1.08 cm/s, respectively.

3.3.3. Settling velocity

The settling velocity of the granules at the end of the adaptation period in R_1 and R_2 were 2.61 and 2.32 cm/s, respectively. Light granules were washed out, and heavier ones with a settling velocity greater than 1.2 cm/s were retained in the reactors. On day 86, the average settling velocity in the R_1 and R_2 were 3 and 2.75 cm/s, respectively. Due to the rapid increase in OLR on day 94,these amounts decreased to 2.62 and 2.44 cm/s. As seen in Fig. 10, from day 94 until the end of the operation, the profile had a monotonic trend and was in the range of 3.12–3.19 cm/s and 2.71-3.01 cm/s by reactor. The greater velocity of the granules in

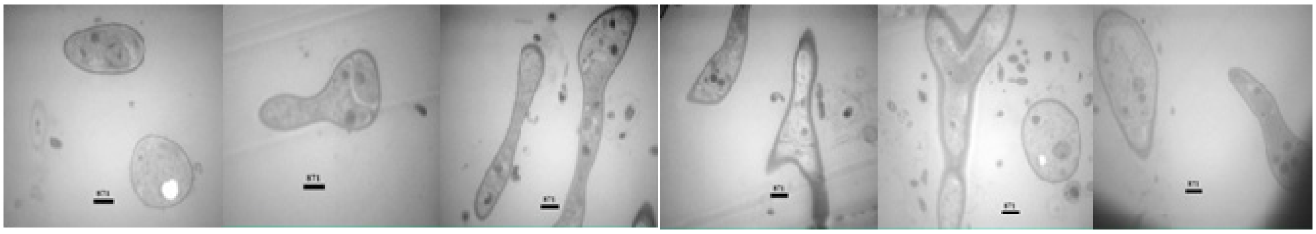

Fig. 8. TEM images of mature aerobic granule after 150 d of operation.

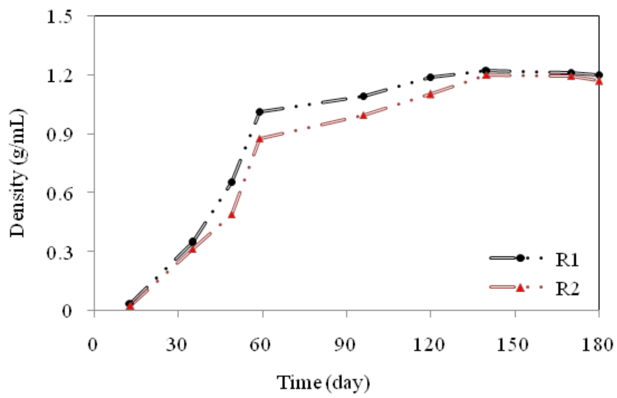

Fig. 9. Variation of granule's density over time in R_1 and R_2.

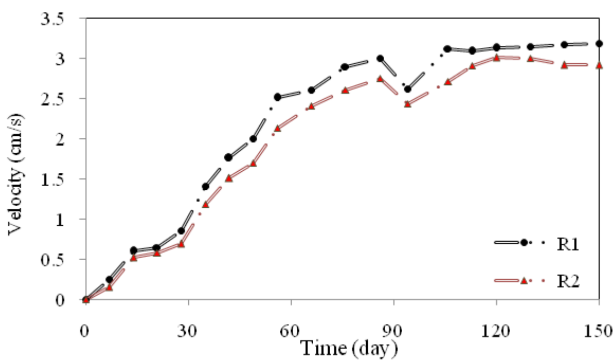

Fig. 10. Variation of settling velocity in bioreactors.

R_1 was due to their larger size compared to those in R_2. This parameter clearly shows the baffles' effect in R_1, because the baffles caused the formation of dense granules with more sedimentation velocity compared to R_2.

3.3.4. Physical strength

Integrity coefficient (IC) evaluated the physical strength of the granules, which indirectly indicated the compactness and activities of the granules. Five samples with different densities were selected to perform this test. Samples with a density equal to 0.0251, 1.0689, and 1.2234 gr/ml, had IC values equal to 54.2%, 21.3%, and 15.4%, respectively. Results can be seen in Fig. 11. Denser and stronger granules in the structure had lower IC. The lower IC value represents the higher granule strength, which results in keeping the

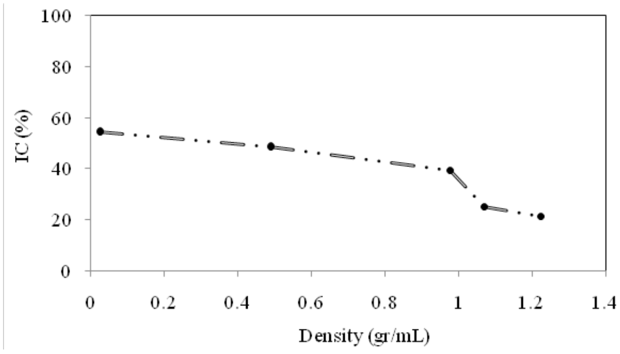

Fig. 11. Profile of IC (%) versus density.

microorganisms together in a granule. It is also noteworthy that permanent entrance of oily wastewater, caused increasement of filamentous genera which could have contributed to granule compaction as they play crucial role in formation of granules.

Rosman et al. [29] report a mean density of 0.0782 g/ml for granules with mean diameter in the range of 2.0 ± 0.1 and settling velocity of 1.69 cm/s.

3.4. GC-MS analysis

Since some compounds typically present themselves in relatively low concentrations, a GC-MS analysis was performed to identify the quality and quantity of the pollutants' different molecules before treatment with SBAR (sample a) and 60 and 360 min after treatment (samples b and c). This analysis was performed in optimal COD_{in} (600 mg/L) at neutral pH, 25°C and 30% humidity. The results show that influent mainly consisted of normal alkanes (C_{14} to C_{28}) and some isoalkanes. Fig. 12 shows the results of chromatography for samples a, b, and c. When comparing the peaks of the charts, it is evident that the concentration of these compounds reduced significantly after six h' treatment, thus confirming the ability of SBR technology in treating petroleum wastewater. For example, compounds such as hexacosane ($C_{26}H_{54}$) (1), heptacosane ($C_{27}H_{56}$) (2), and octacosane ($C_{28}H_{58}$) (3), which had 5.07%, 4.30%, and 2.81% by weight in sample a, were treated completely and became equal to zero in samples b and c. Weight ratio for compounds such as 2-methyl decane, dodecane, and tridecane were equal to zero in samples a and b, but after 360 min, weight ratio for 2-methyl decane($C_{11}H_{24}$) (5), dodecane ($C_{12}H_{26}$) (4), and tridecane ($C_{13}H_{28}$) (6), became equal to 2.12%, 4.69%, and 4.82% in sample c. Generation of these

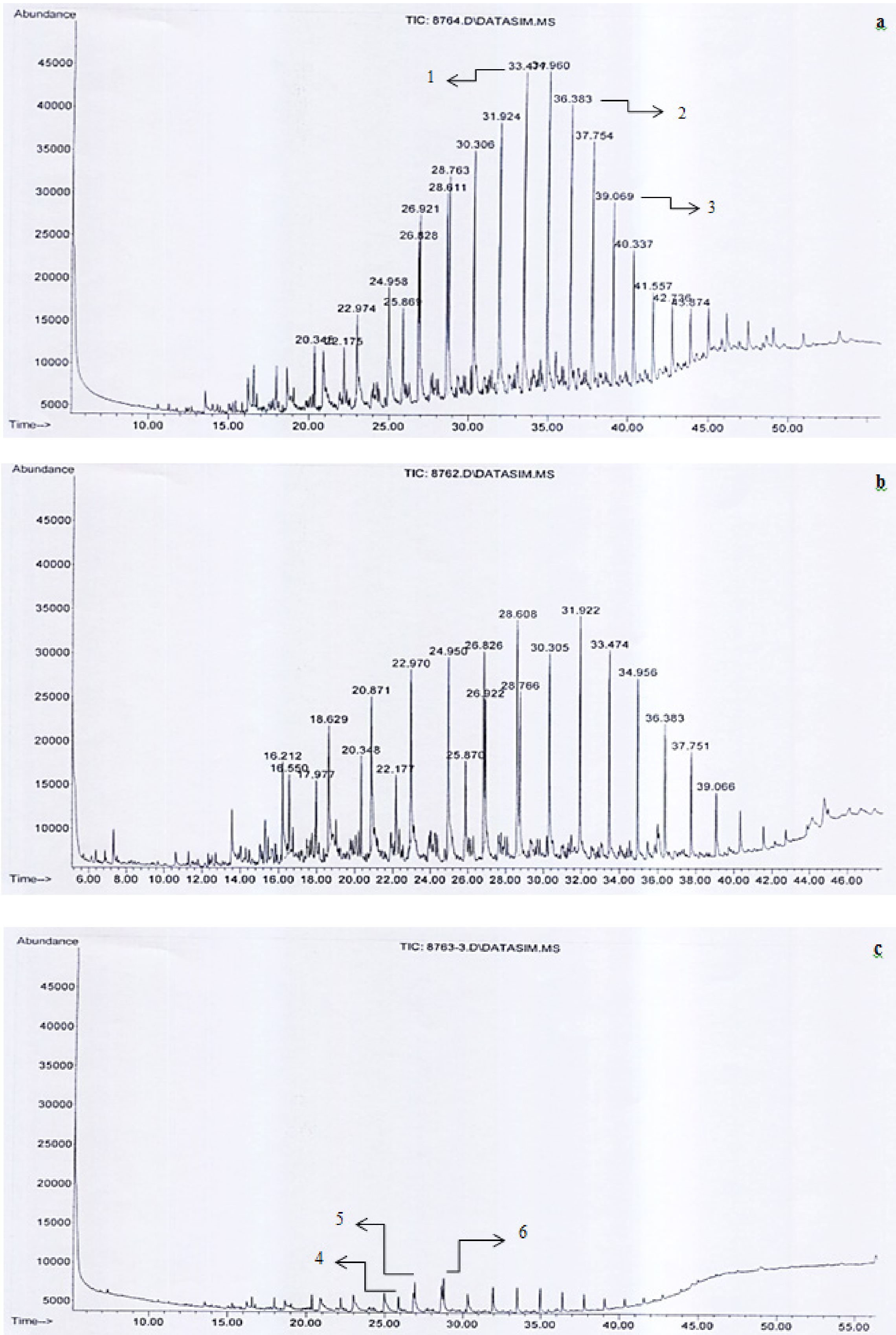

Fig. 12. Chromatogram of oily wastewater.

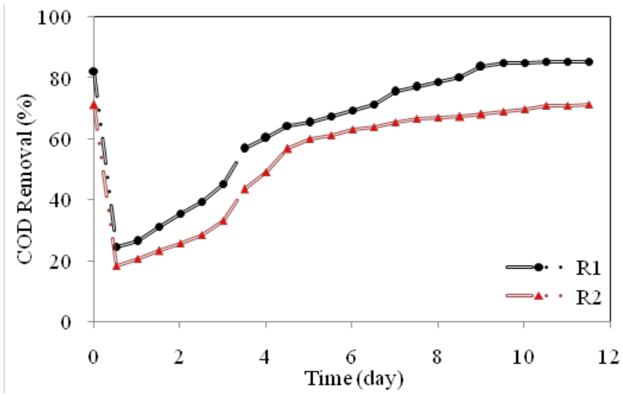

Fig. 13. Performance of reactors and COD removal after shock.

compounds indicated that the heavier alkanes had been broken after 6 h and converted to lighter compounds.

3.5. Effects of the shock on systems operation

With regard to the granules' ability to withstand shock, the OLR in one cycle sharply increased from 1.4 kg/m^{3}d to 1.6 kg/m^{3}d. According to Fig. 13, shock loading significantly decreased the COD removal efficiency in R_1 and R_2 and reduced them from 85.5% and 71.1% to 27.8% and 12.1%, respectively. Results indicated that after 21 cycles, the removal efficiency reached 65.7% and 59%. These amounts reached constant values of 85.2% and 71.2% after 42 and 46 cycles, respectively. R_1 with 57.7% reduction in removal efficiency, returned to the normal condition within 10 days and showed more ability to withstand shock compared to R_2 with 59% reduction in removal efficiency and returning to the normal condition within 11 d.

4. Conclusion

Since R_1 had baffles, sludge traveled further path and rotated more. Therefore, in the same condition, granules formed faster and were larger and denser in R_1 than those in R_2. Moreover, removal efficiency in R_1 was always more than that in R_2. The results also demonstrate the granules' significant physical stability in different conditions. GC-MS results indicated that the concentration of some components have been decreased significantly after six h' treatment, and other heavier compounds have been broken down and have been converted to lighter compounds.

References

[1] W. Khan, I. Najeeb, M. Tuiyebayeva, Z. Makhtayeva, Refinery wastewater degradation with titanium dioxide, zinc oxide, and hydrogen peroxide in a photocatalytic reactor, Process Saf. Environ., 491 (2014) 1–8.

[2] R.C. Selley, S.A. Sonnenberg, Elements of petroleum geology. 3rd ed. Amsterdam: Academic Press, 2014.

[3] J. Jasmine, S. Mukherji, Characterization of oily sludge from a refinery and biodegradability assessment using various hydrocarbon degrading strains and reconstituted consortia, Environ. Manage., 149 (2015) 118–125.

[4] Y. Chang, H.P. Cheng, Sh. H. Lai, G. Ning, Biodegradation of naphthalene in the oil refinery wastewater by enriched activated sludge, Int. Biodeter. Biodegr., 86 (2014) 272–277.

[5] M.H. El-Naas, M.A. Alhaija, S. Al-Zuhair, Evaluation of a three-step process for the treatment of petroleum refinery wastewater, Environ. Chem. Engng., 2 (2014) 56–62.

[6] B.H. Diya'uddeen, S.R., Pouran, A.R.A. Aziz, S.M. Nashwan, W.M.A.W. Daud, M.G. Shaaban, Hybrid of Fenton and sequencing batch reactor for petroleum refinery wastewater treatment, J. Ind. Eng. Chem., 25 (2014) 186–191.

[7] M. Ebrahimi, K. Shams Ashaghi, L. Engel, D. Willershausen, P. Mund, P. Bolduan, P. Czermak, Characterization and application of different ceramic membranes for the oil-field produced water treatment, Desalination, 245 (2009) 533–540.

[8] S. Watcharasing, W. Kongkowit, S. Chavadej, Motor oil removal from water by continuous froth flotation using extended surfactant: Effects of air bubble parameters and surfactant concentration, Sep. Purif. Technol., 70 (2009) 179–189.

[9] M. Hao, Z. Bai, H. Wang, W. Liu, Removal of oil from electric desalting wastewater using centrifugal contactors, Petrol. Sci. Eng., 111 (2013) 37–41.

[10] D. Gao, L. Liua, H. lianga, W.M. Wuc, Comparison of four enhancement strategies for aerobic granulation in sequencing batch reactors, J. Hazard. Mater., 186 (2011) 320–327.

[11] C.E. Santo, V.J.P. Vilar, C.M.S. Botelho, A. Bhatnagar, E. Kumar, R.A.R. Boaventura, Optimization of coagulation-flocculation and flotation parameters for the treatment of a petroleum refinery effluent from a Portuguese plant, J. Chem. Eng., 183 (2012) 117–123.

[12] L. Yan, Y. Wang, J. Li, H. Ma, H. Liu, T. Li, Y. Zhang, Comparative study of different electrochemical methods for petroleum refinery wastewater treatment, Desalination, 341 (2014) 87–93.

[13] T. Misiti, U. Tezel, S.G. Pavlostathis, Fate and effect of naphthenic acids on oil refinery activated sludge wastewater treatment systems, Water Res., 47 (2013) 449–460.

[14] S.O. Rastegar, S.M. Mousavi, S.A. Shojaosadati, S. Sheibani, Optimization of petroleum refinery effluent treatment in a UASB reactor using response surface methodology, J. Hazard. Mater., 197 (2011) 26–32.

[15] N.Sh. Mizzouri, M.d.Gh. Shaaban, Kinetic and hydrodynamic assessment of an aerobic purification system for petroleum refinery wastewater treatment in a continuous Regime, Int. Biodeter. Biodegr., 83 (2013) 1–9.

[16] E.E. Schneider, A.C.F.P. Cerqueira, M. Dezotti, MBBR evaluation for oil refinery wastewater treatment with post-ozonation and BAC for wastewater reuse, Water Sci. Technol., 63(1) (2011) 143–148.

[17] N. Derlon, J. Wagner, R.H.R. da Costa, E. Morgenroth, Formation of aerobic granules for the treatment of real and low-strength municipal wastewater using a sequencing batch reactor operated at constant volume, Water Res., 105 (2016) 341–350.

[18] E. Szabo, M. Hermansson, O. Modin, F. Persson, B.M. Wilen, Effects of wash-Out dynamics on nitrifying bacteria in aerobic granular sludge during start-up at gradually decreased settling time, Water, 8(5) (2016) 172.

[19] B.J. Ni, Formation, characterization and mathematical modeling of the aerobic granular sludge, Springer Theses, 131 (2013) 1–25.

[20] L.K. Wang, Advanced Biological Treatment Processes, Chapter (3): Aerobic Granulation Technology, USA: Lenox Institute of Water Technology; Lenox, MA, 2009, pp.119–124.

[21] SS. Moghaddam, MRA. Moghaddam, Cultivation of aerobic granules under different pre-anaerobic reaction times in sequencing batch reactors. Sep. Purif. Technol., 142 (2015) 149–154.

[22] Y. Song, S. Ishii, L. Rathnayake, T. Ito, S. Satoh, S. Okabe, Development and characterization of the partial nitrification aerobic granules in a sequencing batch airlift reactor, Biores.Technol., 139 (2013) 285–291.

[23] D.J. Lee, Y.Y. Chen, K.Y. Show, C.G. Whiteley, J.H. Tay, Advances in aerobic granule formation and granule stability in the course of storage and reactor operation. Biotechnol. Adv., 28 (2010) 919–934.

[24] S.R. Pajoumshariati, N. Zare, B. Bonakdarpour, Considering membrane sequencing batch reactors for the biological treatment of petroleum refinery wastewaters, J. Membr. Sci., 523 (2017) 542– 550.
[25] S.L. Rezaie, Effect of Chitosan on Granulation Accelerating in GSBAR, Iran, Tarbiat Modaresthesis, 2009.
[26] M.Gh. Rahimi, Total Petroleum Hydrocarbons (TPH) Removal from Wastewater by Using UV/MBBR combined System, Iran, Tarbiat Modares thesis, 2013.
[27] B. Ayati, Determination of mathematical model in treating cellulosic wastewater, Iran, Tarbiat Modares thesis, 2004.
[28] APHA, AWWA, WPCF. Standard Methods for the Examination of Water and wastewater, 20th ed., Washington DC: American Public Health Association Press, (2012).
[29] N.H. Rosman, A.N. Anuar, S. Chelliapan, M.F.M. Din, Z. Ujang, Characteristics and performance of aerobic granular sludge treating rubber wastewater at different hydraulic retention time, Biores. Technol., 161 (2014) 155–161.
[30] J.H. Tay, Q.S. Liu, Y. Liu, Characteristics of aerobic granules grown on glucose and acetate in sequential aerobic sludge blanket reactors, Environ. Technol., 23 (2002) 931–936.
[31] T. Taheri, M.H. Khiadani, M.A. Mohammad, M. Nikaeen, A. Hassanzadeh, Treatment of saline wastewater by a sequencing batch reactor with emphasis on aerobic granule formation, Biores. Technol., 111 (2012) 21–26.
[32] C.J. Tang, P. Zheng, Q. Mahmood, J.W. Chen, Start-up and inhibition analysis of the Anammox process seeded with anaerobic granular sludge, Ind. Microbial Biot., 36(8) (2009) 1093–1100.
[33] Y.M. Liang, Y.L. Yang, Y.W. Chang, J.Y. Chen, Ch. Li, , J.H. Yu, Sh. Chen, Comparison of high pressure and ambient pressure aerobic granulation sequential batch reactor processes, Biores. Technol., 140 (2013) 28–35.
[34] J. Kusmierczak, P. Anielak, L. Rajski, Long-term cultivation of an aerobic granular activated sludge, Electr. J. Pol. Agricult. Univer., 15 (2012) 1–10.
[35] F.A. Dahalan, N. Abdullah, A. Yuzir, G. Olsson, Salmiati, M. Hamdzah, M.F.M. Din, S.A. Ahmad, K.A. Khalil, A. NorAnuar, Z.Z. Noor, Z. Ujang, A proposed aerobic granules size development scheme for aerobic granulation process, Biores. Technol., 181 (2015) 291–296.
[36] Q. Kong, J. Zhang, H.H. Ngo, Sh. Ni, R. Fu, W. Guo, N. Guo, L. Tian, Nitrous oxide emission in an aerobic granulation sequencing batch airlift reactor at ambient temperatures, Int. Biodeter. Biodegr., 85 (2013) 533–538.
[37] Y. Li, J. Zou, L. Zhang, J. Sun, Aerobic granular sludge for simultaneous accumulation of mineral phosphorus and removal of nitrogen via nitrite in wastewater, Biores. Technol., 154 (2014) 178–184.