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Aerobic Granular Sludge: an Advanced Technology to Treat Oil Refinery and Dairy Wastewaters

Abstract- Aerobic granular sludge (AGS) is an emerging and an advance technology for intensive and high-rate biological nutrient removal from wastewater that may play as important role in industrial wastewater treatment strategy in Iraq. Lab scale sequential batch reactor (SBR) operated with selected operation cycle was used to cultivate granular sludge for each of dairy and oil refinery wastewaters. Successful granulation process in each reactor was achieved after 72 day of operation. The results of experimental work proved the flexibility of AGS technology to treat refinery and dairy wastewater and withstand fluctuated load. The COD and NH₄ removal efficiencies for oil refinery wastewater were 86% and 92%, respectively, with sludge volume index (SVI) of 50 ml/g and granules size distribution of (0.3-3) mm. While COD and NH₄ removal efficiencies for dairy wastewater were 80% and 82%, respectively, with SVI of 70 ml/g and the large percentage of its granules were ranged between 0.3 and 0.5 mm and the other smaller percentage were ranged between 0.75 and 2 mm. In addition microbiology observations showed high diverse in microorganisms communities which indicated the presence of stabilized sludge in both reactors.

Key words- Aerobic granular sludge; Sequencing batch reactor; SVI, Settling velocity.

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1. Introduction

Recently, Iraqi water resources have experienced high levels of nutrients, due to effluent discharge from existing wastewater treatment plants. Most of these plants are based on conventional flocculated activated sludge technology and they are suffering from shortness in their capacities, unable to follow the increasing load due population increase and not capable to achieve the required effluent quality. Decentralized treatment of industrial wastewaters is a possible solution to reduce the load on the above-mentioned plants. Many studies were conducted to find advance treatment technologies that reduce the environmental impacts of wastewater and provide the present generation needs [1,2,3]. Aerobic granular sludge (AGS) is an emerging and an advance technology for wastewater biological treatment due to its high sludge settling velocity with best solid liquid separation in comparison with conventional activated sludge system [3,4,5,6]. AGS system is characterized by its small footprint and the removal of organic load and nutrients is achieved in one basin instead of multiple basins in conventional activated sludge system which lowers the construction and the operation $\cos \left[2,7,8\right]$. The distinctive properties of this technology were acquired due to dense and stable structure of its granules [3,7,9]. The compacted structure of these

granules is confirmed as a result of the selfimmobilization of the microorganism that provides good attachment between them and facilitated sludge aggregations [7,10,11]. As well as the selective of specific species of microorganisms occurred during this process that work as back bone for granule structure [12,13,14]. Granulation process can be achieved successfully in sequential batch reactor (SBR) with a selective operation cycle [3,6,8]. In recent years a lot of laboratory scales and pilot studies were conducted for investigating granulation process by using industrial and municipal wastewaters and a number of full-scale plants were built in different places around the world. However to date, there is a lack in data concerning the performance and operation condition of full scale applications, that is because AGS is still in the early stage of expansion and development [9,11,12,15]. This study aimed to investigate the feasibility and flexibility of AGS technology in treating real wastewaters from oil refinery and dairy industries.

2. Materials and Methods

Experimental work on AGS was performed using the pilot scale shown in Figure 1. The system consists of SBR reactor made of glass cylinder with a working volume of 4.42 liter and dimensions of 120 cm length and 7.5 cm

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diameter). Two tanks with volume of 5 liter each were used as influent wastewater storage tanks. The reactor feeding was done by using a pump with flow rate of 100 l/hr. The SBR reactor was aerated by using an air diffuser connected to an air pump. Air flow rate was adjusted by using portable rotameter and a manual control valve. The period of experimental work was 72 days started on 15/11/2014 for each dairy and oil refinery wastewaters.

3. Seeding Sludge Influent and **Compositions**

I. Oil refinery wastewater

The reactor was seeded with flocculated sludge collected from the aeration tank of wastewater treatment system in AL-Dora refinery plant in Baghdad. The seeded activated sludge was filtered using 0.2 mm screen to remove large debris. Oily wastewater, collected from the influent stream of the aeration tank of wastewater treatment system in AL-Dora refinery. The influent composition during the operation period along with the initial mixed liquor suspended solid (MLSS) and SVI of flocculated sludge of the reactor are presented in Table 1.

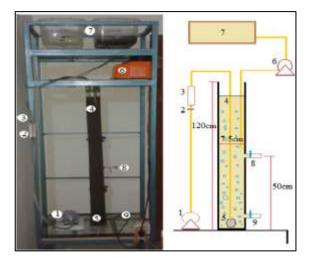


Figure 1: Photo and schematic diagram of pilot scale of AGS. (1.air pump 2. controlling valve 3. air flow meter 4.glass column (120cm*7.5 cm I.D) 5.air diffuser 6.flow pump 7.two tank of wastewater 8. effluent point 9. Sludge sampling point)

Table 1: Influent compositions and initial sludge characteristics of oil refinery wastewater (RR)

Influent composition			Concentration
COD	(mg/l)		92 - 196
TN	(mg/l)		8-17.5
Organic loading rate ORL (g/l. d) Sludge characteristics		0.092 - 0.196	

Initial MLSS	(mg/l)	2850
Initial SVI	(ml/g)	180

II. Dairy wastewater

The reactor was seeded with flocculated sludge collected from MBR system in AL-Zaafrania -Baghdad. The seeded activated sludge was filtered using 0.2 mm screen to remove large debris. Tap water and powdered milk were used to prepare influent synthetic wastewater with the initial sludge characteristics shown in Table 2.

For each of dairy (RD) and oil refinery (RR) wastewater the reactor was operated for a period of 72 days in operation cycle of 12 hr with the same phases consists of 2min for feeding and 11.5 hr for aeration while settling and discharging periods were done at the end of each cycle. Settling period was 30 min for the first 15 days to reduce biomass wash out and then it was reduced gradually to be 5 min at day 30 and to 1 min at day 60 from the beginning of reactor operation. Aeration flow rate in reactor was controlled so that air superficial velocity was about 3.5 cm/sec. The resultant volumetric exchange ratio was 50% per cycle.

COD was measured by using a spectrophotometer (Lovibond). NH4-N and TN were measured by using DR-5000 spectrophotometer (Hatch). Where DO and pН were measured by CX-401multifunction meter (Eutech instrument). MLSS and SVI were measured according to standard methods [17]. The ambient temperature throughout the operation period was in the range 25-30°C. The performance of the system was monitored in terms of wastewater samples quality. The morphology of the flocculated sludge and the aerobic granules for the reactors was observed during operation period using a stereo-microscope and an optical microscope equipped with a digital camera.

Table 2: Influent' components and initial sludge characteristics of dairy wastewater (RD)

Influent composition		Concentration			
Milk powder	(mg/l)	750 -1500			
CH ₃ COONa	(mg/l)	(250) only for the first 10 day of operation period			
NH ₄ Cl	(mg/l)	40			
$\begin{array}{l} K_2HPO_4\\ NaH_2PO_4{\cdot}2H_2O \end{array}$	(mg/l) (mg/l)	27.2 18.7			
TN	(mg/l)	30-66			
COD	(mg/l)	675-1325			
Organic loading (g/L. d) sludge characterist		0.675-1.325			

Initial MLSS	(mg/l)	2540
Initial SVI	(ml/g)	225

After reaching steady state, the microstructure of granules was examined with Scanning Electron Microscope (SEM) at Materials Research Directorate – Ministry of Science and Technology. The size distributions of the aerobic granules were determined by analysis of images taken by a digital camera for random samples of granules groups collected in Petri dishes.

4. Results and Discussion

I. Sludge characteristics

The mixed suspended solid (MLSS) concentration verses operation time for both oil refinery and dairy wastewater reactors are shown in Figure 2. From this figure, it is clear that there was steep decreasing in MLSS concentrations in the both refinery and dairy wastewater reactors (RR and RD) during the first 35 days of operation due to severe biomass washout. The MLSS concentration decreased from 2585 to 1540 mg/l in RR and from 2540 to 1325 mg/l in RD. Then RR presented slight increase in MLSS concentration during the last 25 days due to aggregation of sludge into granules that increased MLSS to 1610 mg/l at the end of operation period. At the same time MLSS concentration in RD was fluctuated between 1340 and 1355 mg/l due to bulking and slow acclimation of biomass to dairy wastewater carbon source (unsteady state). In addition, it has been noticed that sludge color changed from gray to light brown in RR reactor and to light yellow in RD reactor as shown in Figure 3 that can be refer to the differences in wastewater compositions (carbon source) in RR and RD. Granulation process was developed in reactor RR and MLSS concentration improved that produced granules with size range between 0.3 and 3 mm in comparison with reactor RD which showed that the large percentage of its granules range between 0.3 and 0.5 mm and the other smaller percentage between 0.75 and 2 mm as shown in Figure 4 and 5. This might be because RD reactor contained high amount of slowly degradable solid particles of milk that interfere with granulation process and slow it as well as cause the washout of biomass. However the formation of aggregations in the both reactors increases sludge settling velocity and decreases in SVI values during operation period as shown in Figures 6, 7 and 8 which show that maximum settling velocity and minimum SVI value that achieved in RR reactor were 24 m/hr and 50 ml/g, respectively. While, they were 15.5 m/hr and 70 ml/g, respectively in RD. These results indicated that RR reactor present aerobic granular sludge with better performance than RD reactor and that sludge in RD reactor needed more time to integrate its granules. It is important to mention were that Arrojo et al. [11] used real wastewater from dairy product for aerobic granular sludge cultivation with OLR of 7 kg / (m3•d) and then found that granules are formed after 120 day with size of 3.5mm and SVI of 60 ml/g.

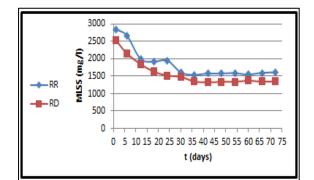


Figure 2: MLSS concentrations in RR and RD reactors verses operation period



Figure 3: Photo of granules of RR and RD reactors

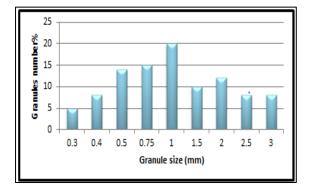


Figure 4: Granules size distribution in RR at the end of operation period

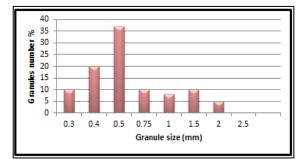


Figure 5: Granules size distribution in RD at the end of operation period

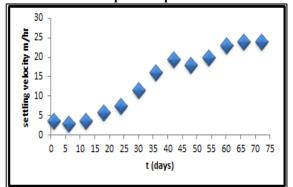


Figure 6: Temporal variation of sludge settling velocity during 72 day of operation in RR reactor

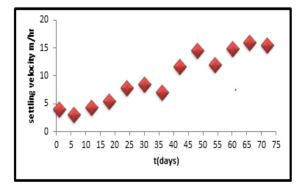


Figure 7: Temporal variation of sludge settling velocity during 72 day of operation in RD reactor

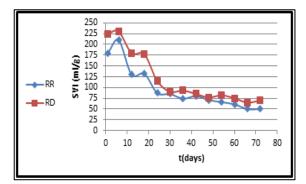


Figure 8: SVI variation in RD and RR during operation period

II. Organic load and nutrients removal

Figures 9, 10 and 11 elucidate the influent, effluent and removal efficiency for COD concentrations of refinery and dairy wastewater in RR and RD reactors, respectively during 72 days of operation period.

These figures show that both reactors RR and RD presented gradual increase in COD removal efficiency as granulation process was developing, so that the maximum of COD removal efficiency in RR and RD in the end of operation period were 86% and 80%, respectively, that corresponded to the effluent COD concentrations of (17.5,265) mg/l respectively. These results indicated that RR and RD reactors succeeded to provide necessary condition for heterotrophic microorganisms growing up. On the other hand the Figures 12and 13 illustrates influent and effluent concentrations of NH4-N in RR and RD which showed that ammonium oxidation process in RR reactor was better than it in RD due to very low influent NH4-N concentrations in RR that facilitated oxidation most of ammonium concentration in comparison with RD reactor, however NH4-N removal efficiency in RR and RD in the end of operation period were 92% and 82% respectively (Figure 14) corresponding to effluent NH4-N concentration of 0.52 and 8.7 mg/l, respectively when the influent concentration of these reactors were 6.5 and 48.8 mg/l, respectively. These results could be an indicator to the activity of ammonium oxidizer in these reactors that they were not affected by the type of wastewater. In contrast with the influent and effluent results of TN(total nitrogen) for RR and RD reactors illustrated in Figures 15 and 16, which indicated that granules size distribution in these reactors may be played the main factor that effected on the results of TN removal efficiency as shown in Figure 17. So that large granule size in RR reactor could provide anoxic condition in their core and encouraged the denitrification process toward removal efficiency of 81%. While the small size of the most granules in RD reactor weaken the nitrogen removal efficiency to 64%. Schwarzenbeck et al [18] achieved 90% of the COD removal efficiency when he used AGS technology to treat effluent of dairy industry with OLR of 5.2 g/l d.

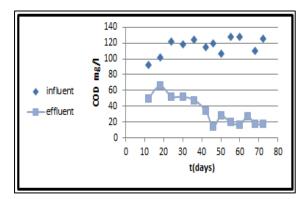


Figure 9: Influent and effluent COD concentration in RR reactor during 72 days of operation

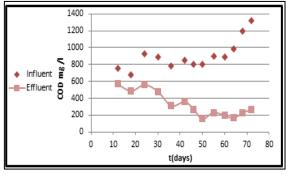


Figure 10: Influent and effluent COD concentration in RD reactor during 72 days of operation

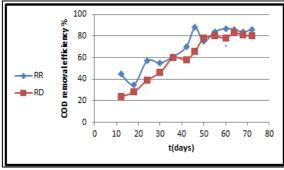


Figure 11: COD removal efficiency in RR and RD reactors during 72days of operation

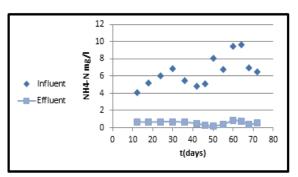


Figure 12: Influent and effluent NH4-N concentration in RR reactor during 72 days of operation

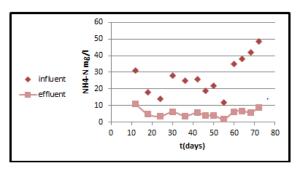


Figure 13: Influent and effluent NH4-N concentration in RD reactor during 72 days of operation

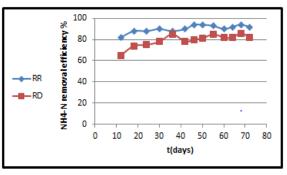


Figure 14: TN removal efficiency in RR and RD reactors during 72 days of operation

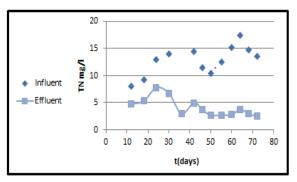


Figure 15: Influent and effluent TN concentration in RR reactor during 72 days of operation

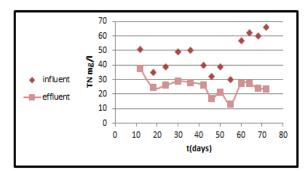


Figure 16: Influent and effluent TN concentration in RD reactor during 72 days of operation

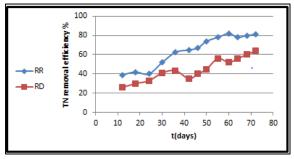


Figure 17: TN removal efficiency in RR and RD reactors during 72 days of operation

III. Microbiology observation

The use of two different wastewaters in this study presented high diverse in microorganisms communities as shown in Figures 18-1 and 19-1. These Figures show that granules surfaces were covered by different type of protozoa, ciliate and rotifers which indicated that the sludge was stabilizing in the both reactors. As well as SEM images (Figures18-2, and 19-2) presented that most of these granules were consist of large percent of staphylococcus bacteria bonded by EPS. Active microbial population on the granules surfaces with the biofilm matrix explained the utilization and removing of organic material in RR and RD reactors.

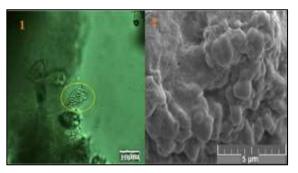


Figure 18: (1) Optical microscope and (2) SEM images of a granule surface in refinery wastewater reactor (RR)

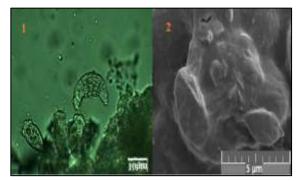


Figure 19: (1) Optical microscope and (2) SEM images of a granule surface in dairy wastewater reactor (RD)

5. Conclusions

The results of this study indicated that AGS technology succeeded to treat oil refinery and dairy wastewater by achieving:

1. 86% and 92%, in COD and NH4 removal efficiencies respectively for oil refinery wastewater.

2. 80% and 82% in COD and NH4 removal efficiencies respectively for dairy wastewater.

3. Good sludge performance for each wastewater was presented by high settling velocity and minimum SVI, which were (24 m/hr, 50 ml/g, for oil refinery wastewater and 15.5 m/hr, 70 ml/g, for dairy wastewater.

4. Granules morphology, diversity of parasites grown on the granule surface as seen by optical microscope and bacteria clustering and EPS as shown by SEM images presented a clear picture of granules activity.

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