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Aerobic Granular Sludge for Leachate Treatment

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The treatment of municipal landfill leachate by means of aerobic granular sequencing batch reactors (GSBRs) was investigated. The paper reports the results from an experimental campaign lasted 100 days, which has been divided into three periods: cultivation of granular sludge (70 days), operation with semi-fresh (15 day) and diluted landfill leachate (15 day). Two different GSBR configurations were used: a Sequencing Batch Bubble Column reactor and a Sequencing Batch Airlift Reactor. All reactors were operated at Volume Loading Rates (VLRs) between 4.8 and 7.2 $g_{COD}/(m^3 \cdot d)$. The Chemical Oxygen Demand (COD) removal efficiency varied between 80% and 90% under operation with synthetic wastewater feeding. On the other hand, the COD removal performance decreased to 40-50 % with semi-fresh leachate and to 50-60% with diluted leachate. Regarding nitrogen removal, after granules formation, the performance were satisfactory only when the reactors were fed with synthetic wastewater. Contrarily, the obtained results underline that a specific pre-treatment of ammonium must be applied in order to optimize nitrogen removal. However, the observed results indicate that the landfill leachate can be potentially treated in GSBR bioreactors.

1. Introduction

Aerobic granulation is a new environmental biotechnology for treating a wide variety of wastewater (among others Adav et al., 2008). Indeed, this technology has shown a great potential in wastewater treatment due to several advantages represented by excellent biomass settle-ability, high biomass retention, good ability to treat high organic loading rate (Liu and Tay, 2006a). The aerobic granular sludge can be easily obtained in sequencing batch reactors with different configuration. In particular, the mostly used Granular Sequencing Batch Reactor (GSBR) configurations are: Sequencing Batch Bubble Column reactor (SBBC) and Sequencing Batch Airlift Reactor (SBAR) (Beun et al., 2002).

Generally, the operational conditions imposed by a sequential procedure allow to operate a selective process between fast and slow settling biomass. Consequently, due to bacteria self-immobilization, the selected biological aggregates will gradually change in real stable granules (Liu and Tay, 2002). This initial selection phase can be called "cultivation phase" and it is considered as a physical screening step (or physical settling-washing out action) that operates a "selection pressure" on the biomass in the reactor (Liu and Tay, 2006b). Thus, only the aggregate that becomes big and dense enough to quickly settle would be retained in the reactor. In general, the cultivation phase is carried out gradually decreasing the settling time-length, in order to improve the selection pressure efficiency (Torregrossa et al., 2007).

Granules formed in such a way are compact and characterized by an outer spherical shape. Further, the structure of stable granule is characterized by different layers: in the inner part of the granules, it can be observed the presence of heterotrophic population and, due to oxygen diffusion limitation inside the granules, it is possible to establish a denitrification process; in the intermediate layer, autotrophic biomass is dominant; in the outer layer, where oxygen and organic substances are highly available, heterotrophic growth occurs (Jin et al., 2008). The correct development of granulation, and consequently the actual granule stratification, depends on some important factors and conditions (Kim et al., 2008): feast and famine alternation, related to the duration of external substrates feeding; duration of the starvation phase; superficial air velocity; hydrophobic condition of the mixed liquor; organic loading rate. For these reasons, the granules can be formed after an unspecified time. Although aerobic granulation in GSBR has been extensively investigated, most of the previous studies concerning the cultivation of aerobic granules have been carried out with synthetic wastewater (Wey et al., 2012). Only few studies with real wastewater are available to better characterize the aerobic granulation process with domestic sewage (Kim et al., 2008; Ni

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et al., 2009). Reports of successful aerobic granulation systems were also obtained with other real readily biodegradable wastewater such as dairy effluent, malting wastewater, brewery wastewater (Schwarzenbeck et al., 2004 and 2005). Contrarily, the treatment of toxic and refractory wastewater is still lacking (Wey et al., 2012). In this context, the paper reports a case study that describes the organic carbon and nitrogen removal during cultivation period with synthetic wastewater in aerobic granular sludge SBR, and a subsequent treatment of landfill leachate at different organic and ammonium loading rates.

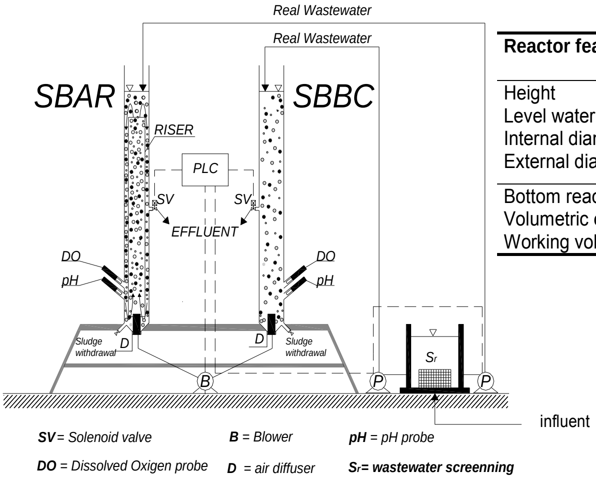
2. Materials and Methods

2.1 Experimental set-up

Two different reactors, named R1 and R2, were used. In particular, the entire experimental period lasted 100 days, which has been divided into three sub-periods:

- Period I (70 days) - cultivation of granular sludge with synthetic wastewater;
- Period II (15 day) - operation with "semi-fresh" leachate;
- Period III (15 day) - operation with leachate "strongly diluted" with synthetic wastewater.

During Period 1 SBBC and SBAR configuration were used. Contrarily, during Periods II and III, both reactors were operated under SBBC configuration. All the reactors, were constituted by a working volume of 3.5 L. Other features and the scheme of experimental installation are reported in Figure 1.



Reactor features	Units	SBBC	SBAR	
			External cylinder	Riser
Height	mm	860	860	600
Level water table	mm	700	740	740
Internal diameter	mm	80	80	54
External diameter	mm	90	90	60
Bottom reactor – riser distance	mm		30	
Volumetric exchange ratio	%	50	50	
Working volume	L	3.5	3.5	

Figure 1 *Pilot plant layout: geometric characteristics of reactors*

The average characteristics of synthetic wastewater (Period I) and leachate (Period II and III) are shown in Table 1. In particular, leachate was collected from Palermo Municipal landfill. Preliminarily, aged and fresh leachates were mixed and stored, in order to maintain similar characteristics during the whole experimentation. Subsequently, during Period II, it was used leachate with a pollutants concentration greater than ones in the Period III, in order to study the potential occurrence of granule breaking effect due to the different nature of the substrates in the influent (after initial cultivation). More specifically, in order to obtain a "semi-fresh" mixture a weak dilution (with tap water) was applied (in order to maintain an average concentration of 9600 mg/L). Contrarily, in Period III, in order to evaluate the possible improvement of system performance, and the eventual new granulation, a "strong" dilution of leachate was applied (until reaching a average COD concentration of 4800 mg/L). The synthetic wastewater was only used during cultivation and in R2 during period II. This last condition was applied in order to compare the performance of two reactors fed with different wastewaters, maintaining the same Volumetric Loading Rate (VLR). The plant was started-up with an initial settlement time (t_S) of 7 minutes that was decreased to 3 minutes after only 2 weeks (Torregrossa et al., 2007). The cyclic operation of SBR has been automated using a programmable logic controller (PLC). In order to change the VLR during the experimentation, without changing the COD concentration in the wastewater, the cycle time was changed (generally, longer times were applied when feeding leachate, according to Wey et al., (2012)). More specifically, in order to balance the VLR, the reactors have been operated as follows: 8 daily cycles of 3 h each, during cultivation (Period I) and when synthetic wastewater was fed in R2; 1 daily cycles of 24 hours and 3 cycles of 8 hours when "semi-fresh" or "diluted" leachate were fed, respectively. In table 2, all operational conditions during the whole experimentation are summarized.

Table. 1. *Composition of synthetic and landfill leachate fed to the GSBRs*

	Parameter	Period I		Period II		Period III	
	[mg/L]	R1$_{(SBAR)}$	R2$_{(SBBC)}$	R1$_{(SBBC)}$	R2$_{(SBBC)}$	R1$_{(SBBC)}$	R2$_{(SBBC)}$
Syntetic wastewater	**COD**	598 ± 50	598 ± 50		1185 ± 101		
	COD$_{sol}$	162 ± 13	162 ± 13		340 ± 24		
	N-NH$_4$	25 ± 2.4	25 ± 2.4		51 ± 8.1		
	N-NO$_3$	0.3 ± 0.1	0.3 ± 0.1		0.3 ± 0.1		
	N-N$_{tot}$	30 ± 2.4	30 ± 2.4		55 ± 9.4		
Leachate Weak diliution	**COD**			9738 ± 450			
	COD$_{sol}$			2800 ± 125			
	N-NH$_4$			1960 ± 82			
	N-NO$_3$			0.5 ± 0.4			
	N-N$_{tot}$			3700 ± 285			
Leachate Strong diliution	**COD**					4560 ± 165	4560 ± 165
	COD$_{sol}$					1540 ± 175	1540 ± 175
	N-NH$_4$					945 ± 54	945 ± 54
	N-NO$_3$					0.3 ± 0.4	0.3 ± 0.4
	N-N$_{tot}$					1845 ± 175	1845 ± 175

Table. 2. *Operational condition during the period I, II and III.*

	Experimental period	**Period duration**	**Range**	**Type**	**Cycle**	**Duration of single SBR phase**				**VLR**
						t_F	**t_{AIR}**	**t_S**	**t_{DS}**	
		days	days	h	min	min	min	min	min	kg$_{COD}$·m^{-3}·d^{-1}
R1	I Synthetic wastewater	70	1-7	SBAR	180	5	163	7	5	2.4
			8-14	SBAR	180	5	165	5	5	2.4
			15-21	SBAR	180	5	167	3	5	2.4
	II Leachate Weak dil.	15	71-85	SBBC	1440	5	1427	3	5	4.8
	III Leachate Strong dil.	15	86-100	SBBC	720	5	707	3	5	4.8
R2	I Cultivation	70	1-7	SBBC	180	5	163	7	5	2.4
			8-14	SBBC	180	5	165	5	5	2.4
			15-21	SBBC	180	5	167	3	5	2.4
	II Synthetic wastewater	15	71-85	SBBC	180	5	167	3	5	4.8
	III Leachate Strong dil.	15	86-100	SBBC	480	5	467	3	5	7.2

t_F – Fill time; **t_{AIR}** – Aeration time; **t_S** – Settling time; **t_F** – Fill time; **t_{DS}** – effluent discharge time

2.2 Analytical procedure

In both experimental periods, the chemical-physical parameters analysed were: total COD, soluble COD (COD$_{sol}$), total suspended solids (TSS), volatile suspended solids (VSS), ammonium nitrogen (NH$_4$-N) and nitrate nitrogen (NO$_3$-N), with regards to influent, mixed liquor and effluent. In particular, the biomass concentration (in term of TSS), in the reactor and in the effluent, has been determined by filtrating the homogeneous sample using a 0.45 μm filter and drying the filter for at least 24 h at 105°C. All the other chemical-physical analyses have been carried out according to the Standard Methods [APHA, 1998]. Sludge characteristics have been analyzed by means of microscopic and stereoscopic observations. In particular, microbiological analysis have been carried every two-three days. More specifically, a sample of granular sludge (50 mL) has been analysed through a stereoscope that acquired the microscopic images, using an opportune enlargement (relating to granule sizes). The images has been analysed by a specialized IA (IM50 by Leica). It is important to specify that the granule dimensions have not been evaluated through a statistical measure, but rather as average value of the maximum dimensions observed in the mixed liquor sample.

3. Results

3.1 Cultivation Period

The cultivation phase was aimed to the granular sludge production: for this reason, as discussed above, the plant was fed with synthetic wastewater. By analyzing the granulation phenomenon as well as on the basis of the large number of measurements, some interesting aspects can be drawn. In particular, as underlined by the average data shown in Figure 2, the reactor R1 in SBAR configuration, showed a slightly better granulation phenomenon compared to that one occurred in the reactor R2 in SBBC configuration. Briefly:

1. the removal of the organic substance (in terms of COD) was satisfactory in both reactors and improved with the growth of biomass, reaching values higher than 85%, at the end of Period I;
2. nitrification (N) occurred efficiently only when the granules reached an average size greater than 0.5-0.6 mm (after the 22nd day in R1 and after the 29th day in R2);
3. denitrification (DN) have been particularly affected by the growth and stratification of the granules and only at the end of Period I the best results have been achieved: in any case, the denitrification was

always greater in the granules of reactor R1 (SBAR), because they were larger and (obviously) better stratified than granules in R2.

	Cultivation Period							
	from	to	ηCOD	ηN	ηDN	size	TSS	EPS_{tot}
	day	day	%	%	%	mm	g/L	mg/gVSS
R1	0	7	77.7592	53.0259	15.5584	0	9.55933	113.251
	8	14	80.602	64.5907	25.9756	0.2	13.547	142.674
	15	21	84.6455	68.1376	17.3247	0.39	13.687	213.892
	22	28	87.9462	72.4552	42.1735	0.5	19.1565	337.409
	29	49	89.8198	76.436	63.0129	0.63667	23.0181	271.599
	50	70	91.0502	77.2789	81.4889	0.93	17.1748	228.116
R2	0	7	76.087	46.2048	8.1319	0	8.18219	71.9758
	8	14	79.097	57.7362	21.1685	0.1	19.4384	219.625
	15	21	83.1104	64.4001	25.0222	0.32	18.1848	293.365
	22	28	86.9565	68.9198	33.924	0.44	14.4807	252.658
	29	49	87.3416	73.7428	50.7918	0.55333	18.2208	311.917
	50	70	88.709	76.036	70.7455	0.85667	22.55	261.141

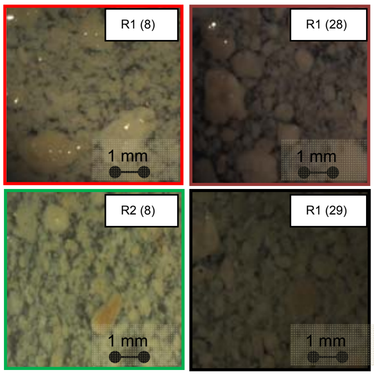


Figure 2 *Granule sizes during cultivation phase*

Therefore, the observed results confirmed the usefulness of the airlift in the column during the cultivation phase (Di Bella and Torregrossa, 2013), since a more regular shear stress can be obtained. Moreover, it appeared that the mechanical stress, due to the presence of the airlift, initially caused a greater production of EPS_T. The latter acts as "glue" for other biological aggregates, facilitating the initial formation and growth of granules. Unfortunately, the analysis of EPS_T was not performed in the subsequent periods, due to the influence of leachate compounds in the colorimetric analysis.

3.2 Experience with leacheate

As previously discussed, starting from Period II, both reactors were configured as SBBC (by removing the airlift in R1). Furthermore, in order to properly compare the reactors, the granular sludge of R1 (the "best" one) was redistributed in both reactors with a starting concentration of about 11 g/L in terms of TSS. More specifically, only a portion of granular sludge of R1 was collected (after a preliminary selection of the granules with best settle-ability). The only differences were: the type of wastewater (leachate or synthetic) and the operating conditions (VLR), as already shown in the table 2.

3.2.1. Granulation

The available studies on granulation in GSBR systems fed with landfill leachate are very limited, especially under aerobic conditions. The study of Wei et al. (2012), already cited in this paper, was based on the experimental observation of the performance achieved thanks to a start-up period carried out directly with leachate: although useful, this choice has limited the size of the granules to values lower than 0.5 mm. On the contrary, in the present experience, the evolution of the granulation resulting by organic and ammonium shock loads has been studied: starting from an ideal environment for the granules growth (synthetic wastewater), it was "abruptly" fed leachate (more or less diluted). The aim was to study what happened to the granules. Figure 2 shows the trend of the average size of the granules and the concentration of the biomass (in terms of SS).

As shown in Fig. 2a (Period II) the average granule sizes tended, as expected, to decrease when the leachate was fed to the system. On the contrary, in the R2 (Fig 2b) the granule sizes were maintained (apart from the immediate shock due to the jump of VLR, from 2.4 to 4.8 kg COD/(m^3 d)) and even increased at the end of Period II. From day 85 (Period III), the granulation showed a different trend: in the reactor R1 there was a formation of new granules and the unstructured ones have begun to "grow"; while in R2 the granules have begun to break down. This was probably due to the fact that the leachate was fed in R1 when the biomass was partially acclimated, but with concentrations of leachate lower than ones in the Period I (maintaining the value of VLR, via the reduction of the cycle from 24 to 12 hours). On the contrary, in reactor R2, in Period III it was fed real leachate to the biomass previously acclimatized with synthetic wastewater (low concentrations but at a higher load, from 4.8 to 7.2 kg COD/(m^3 d), with 3 cycles of 8 hours daily). However, despite the high stress imposed, the granule breaking was limited and less evident, probably because the granulation and stratification were more efficient.

Obviously, the SS concentration trends in the reactors were influenced by the rupture of the granules and cyclical conditions of "washout". This, of course, had obvious repercussions on the performance of the process, as will be shown below.

3.2.2. COD and ammonium removal

The performances, in terms of COD removal, are shown in Fig 3.

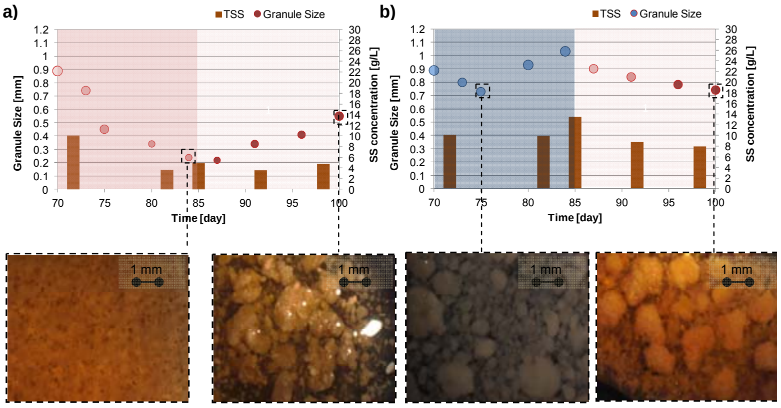


Figure 3 *Granule sizes and TSS concentration in R1 (a) and R2 (b)*

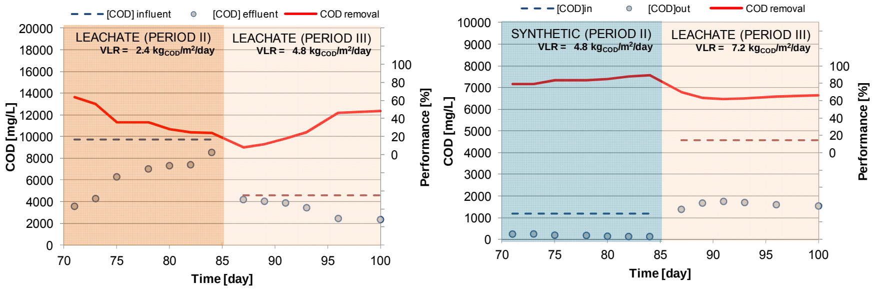


Figure 4 *Organic removal performance*

It is important to underline that the experimentation was carried-out under dynamic conditions during Period II and III. However, the results showed that a stable organic was achieved in both reactors, but the COD removals are meanly low: 45-50% in R1 and 60-65 % in R2. In fact, the organic removal was strongly dependent on the changing of influent, from synthetic to leachate. Only in R2 during Period II, with synthetic wastewater, the COD removal are very good, always higher than 80% despite the high COD concentration (about 1200 g/L). Regarding the leachate feeding in Period III, according to observation reported above, the data show that the granulation phenomenon start again in R1 when leachate was fed. Summarizing, from the 70th to 85th day of R1 the removal efficiency decreased from 70% to 20% due to strong defflocculation of the granules (and washout of biomass). In the same period, R2 was fed with synthetic wastewater with a COD concentration of 1185 ± 105 mg/L and COD removal fluctuated in the range 78-88%. In Period III, the COD removal in R1 increased reaching a value equal to 45%, thanks to the establishment of a new phenomenon of granulation. Contrarily, the COD removal in R2 decreased but only to 55%, despite the high organic loading rate and the changing of the influent (from synthetic to leachate). On the other hand, Figure 3 shows the results in terms of Nitrification (N) and Denitrification (DN) performance in both reactors. In general, biological nitrogen removal normally involves two separated step: aerobic nitrification of ammonium to nitrate; anoxic denitrification of nitrate to nitrogen gas. In the granular sludge process the simultaneous nitrification and denitrification (SND) is generally developed (Di Bella and Torregrossa, 2013). In this context, the denitrification-nitrification activity depends on granule size because, without dissolved oxygen control, the oxygen penetration change with granules diameter. In particular, the SND process is satisfactory when synthetic wastewater was fed: both denitrification and nitrification performance higher than 70-80% (data are not reported). On the other hand, when the leachate was used as influent, the nitrogen conversion occurred at "*high ammonium concentration*": in this condition, according to Wey et al., (2012), *nitrifying bacteria in aerobic granules was enriched, while the heterotrophs lost their competitive dominance, which anticipated the simultaneous ammonium oxidation and organic biodegradation.* Besides this aspect, due to ammonium oxidation production, the activities of both ammonium oxidizing bacteria (AOB) were inhibited, resulting in the accumulation of ammonium (lower nitrification efficiency) in both typical cycles. Nitrate concentration was also increasing due to decrease in denitrifying activity. For this reason, the denitrification performance resulted not satisfactory (<20% in R1 and <40% in R2), despite the granular sizes are not very small. this result was similar to what observed for nitrification. Probably, for leachate treatment in GSBR, a specific pre-treatment for ammonium removal must be applied, in order to reduce the ammonium concentration in the effluent.

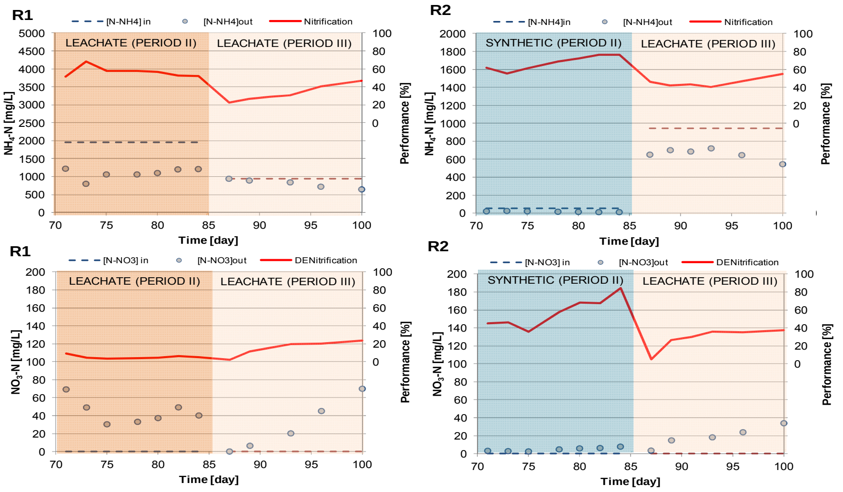


Figure 5 *Nitrogen removal performance*

4. Conclusion

The aim of this work has been to observe the phenomenon of the aerobic granulation in two GSBR fed with landfill leachate The result confirm the possibility to obtain aerobic granule size feeding landfill leacheate. However, this study has confirmed the results reported in the literature highlighting the decisive role of ammonium concentration: in particular, the organic and nitrogen removal tend to decrease when ammonium concentration increase. For this reason, a specific ammonium pre-treatment must be applied before aerobic granular sludge application. Furthermore, the initial cultivation of granular should be carried out with small increment of lecheate, in order to optimize the bacteria acclimatation and specification.

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