Journal of Civil & Environmental Engineering

Drakides and Lay-Son, J Civil Environment Engg 2012, 2:3
DOI: 10.4172/2165-784X.1000112

Research Article | Open Access

Aerobic Granules Formation in Dual-Layer Percolating Filters

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Abstract

Aerobic spherical granules were observed in zeolitic dual-layer intermittent percolating filters treating domestic septic tank effluent. The diameter of the granules ranged from 0.05 to 0.5 mm and when they predominate in the biofilms the mean SVI value was of 54 ml g^{-1} instead of 232 ml g^{-1} when they were absent.

Flow behavior in these filters was characterized by the dimensionless Reynolds number. The values obtained ($40<Re<60$) were representative of intermediate flow conditions and invalidated the current hypothesis of laminar flow conditions. These values were of the magnitude of those estimated from studies on SBR reactors presenting granular sludge formation.

The possible influence of granular sludge formation on long-term performances and interfacial clogging of intermittent dual-layer percolating filters is also discussed.

Keywords: Aerobic spherical granules; Reynolds number; Percolating filters; Waste water treatment

Introduction

Aerobic spherical granules (ASG) formation is considered as a special case of biofilm growth. It was reported first in an aerobic upflow sludge blanket (AUSB) reactor, but it was mainly reported in sequencing batch reactors (SBR) [1]. The cyclic operation of SBR consisting in influent filling, aeration, settling and effluent removal is considered to enhance ASG formation [2].

Reported diameters for aerobic granules range from 0.25 mm for industrial wastewater [3] to 2.6 cm for synthetic wastewater, with excellent settleability [2].

Many operational parameters have been reported to influence ASG formation [4-8,2], but it must be noted that most of experiments are based upon synthetic (acetate, molasses, sucrose, etc) or industrial wastewaters: dairy wastewater [9], soybean-processing wastewater [3], brewery wastewater [10], fish canning wastewater [11]. Cultivation cases of ASG for the treatment of domestic wastewaters are much more limited, De Kreuk et al. [12] reports heterogeneous aerobic granular structures formation after 20 days of operation in a SBR.

Hydrodynamic shear forces, commonly linked in SBR with aeration intensity (air superficial velocity), are associated very closely to ASG formation, and within a given operational range, help to stabilize the three dimensional structure of the granules and enhance microbial granulation process [4,7]. Nevertheless, the mechanism by which these forces influence formation of granules is still not well understood.

Up to now, few reports are available of ASG formation in wastewater treatment reactors others than SBR. Di Iaconi et al. [13] observe biomass granulation during the start-up period of a sequencing batch aerated biofilter and Beun et al. [14] shows that aerobic granular sludge can easily be cultivated in a sequencing batch airlift reactor. Particularly, no case is reported of ASG formation in static saturated or percolating filters.

On another side, percolating filters have been extensively studied, particularly for the treatment of domestic wastewaters following septic tank pre-treatment [15]. Intermittent filtration is well-known to allow an increase of the filters loading rate whilst avoiding an early clogging, particularly through fractioning the daily load in several flushes [16]. Most of studies make the implicit assumption of laminar flow conditions, under which diffusion processes control the degradation of dissolved organic contaminants by biofilms fixed upon the filtering media. Performances are then linked to the specific media superficy, the biofilm thickness, and the hydraulic retention time and diffusivity coefficients [15].

Systems for household wastewaters treatment in France include currently the well-known sand intermittent percolating filters, but also a specific application of dual-layer percolating filters (DLPFs) using natural zeolites of defined granulometry as filtering media (French 2009-9-7 Application Decree of the European Normalization Project EN 12566-6 - PrEN). These later systems have not been extensively studied.

In this work we report the steady-state formation of ASG in experimental DLPFs zeolitic systems treating domestic septic tank effluents. The Reynolds number Re, appears as the key factor affecting biofilm granulation in such systems, allowing to introduce an eddy flow concept in place of the current laminar flow hypothesis. The relationship between ASG formation and filters clogging, key factor for the long term stability of DLPFs, is also discussed.

Material and Methods

Percolating filters

Six columns of transparent PVC with 15 cm diameter and 90 cm high were installed and filled according to the above-mentioned Decree. One column was filled with 70 cm of a sand of prescribed granulometry as a reference filter, and the others with 55 cm of natural zeolitics materials, one chabazite (reference ZN 024-14) and two clinoptilolites (clino 1 and clino 2 with the references ZN 324B-08 and ZN 324-24 respectively) provided by SOMEZ Inc (St Jean de Védas, France). The zeolitics materials columns were filled with two layers

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Received December 20, 2011; **Accepted** March 29, 2012; **Published** March 31, 2012

Citation: Drakides C, Lay-Son M (2012) Aerobic Granules Formation in Dual-Layer Percolating Filters. J Civil Environment Engg 2:112. doi:10.4172/2165-784X.1000112

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of different granulometries, 2-5 mm at the top and 0.5-2 mm at the bottom for chabazite and clino 1 and 2-5 mm at the top and 1-2 mm or 0.3-0.7 mm at the bottom for clino 2. The height of each layer was 40 cm for the top layer and 15 cm for the bottom one. All fillings were washed prior to use.

Fillings granulometrics characteristics are given Table 1, with d_{10} is the maximum diameter of the 10% finest fraction, (d_{60} for the 60%) and Uc (uniformity coefficient) = d_{60}/d_{10}.

For each column, a peristaltic pump controlled by a clock feeded a tilting bucket fitted with an electronic switch, delivering a constant volume that could be adjusted between 0.1 and 0.3 L. A ten cm layer of 1.5-2.5 cm diameter gravel was installed at the top of the column, in order to spread evenly the water on the column surface. As a draining layer, fifteen cm of the same gravel were installed at the bottom of the column and separated from the above layer by a 1 mm mesh geogrid. The diagram of the experimental columns is shown in Figure 1.

Wastewater characteristics

The wastewater was collected from a septic tank of 3 m^3 volume providing a 5 days hydraulic retention time and fed with domestic wastewaters originating from the village of St Clément-de-Rivière (France). The septic tank effluent had the following analytical mean values as mg l^{-1} (min/max): BOD_5 = 173 (56/301); COD = 395 (169/708); TSS = 195 (36/527); NH_4 as N = 88 (20/200); PO_4 as P = 6.6 (1.3/12.8). These values lay within the range recommended by the PrEN.

Operating conditions

Feeding was divided in seven flushes per day reproducing a daily flow pattern in accordance with PrEN, with the exception of the sand filter that received only four flushes. The considered flow rate was 600 l d^{-1} for a housing corresponding to 4 inhabitants. This flow was considered applied on the reference surface given for each filling by the Decree, 25 m^2 for the sand filter (C1), 5 m^2 for the zeolitic filters (C2, C3, C4-1, C5), and 10 m^2 for (C4-2). The column number 4-2 was used to test a smaller loading rate.

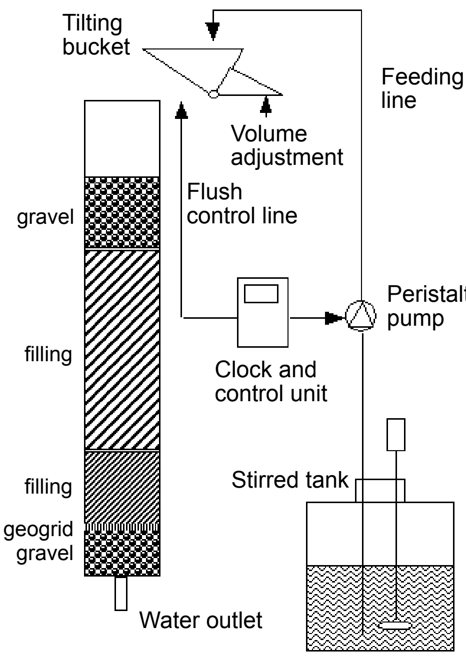

Figure 1: Diagram of the experimental columns.

Resulting applied surfacic loads as well as the numbers and the volumes of flushes are shown in Table 2. These loads proved to be close from those used in other studies on intermittent filtration [16,17].

Instead of the 44 weeks follow-up asked by the PrEN, the columns were feeded for more than two years (123 weeks). Performances and permeability of systems went on evolving during the experiment, and clogging occurred for the chabazite C2 and the smaller sized clinoptilolite C5 after the 84th week (Table 2).

Analytical methods

Microbial observations were conducted with a photonic microscope Olympus BX 50 (Japan) fitted with a Burker counting chamber (Assistent, Germany).

For each column, we observed effluent samples at different dates and biofilms extracted from the fillings sampled at different heights during the dismantling of the columns at the end of the experiment. Extracted biofilms were obtained by washing gently 100 ml of filling with treated water. Biofilms decanted readily and were recuperated in 100 ml fractions. Representative 0.1 ml subsamples were placed in the Burker counting chamber for direct microscopic examination. ASG density was evaluated in four categories: absent, present but scarce (< 10% of total biofilm volume), present (10% < percent < 20%), and abundant (>20% and up to 50%).

Aliquots of the same suspensions were filtered through ash-free paper filters (Durieux n°111, France) prior drying at 105°C and burning at 525°C to determine gravimetrically TSS and VSS (balance Precisa XT 120A). The obtained VSS was reported to the layer volume and used to evaluate the total volatile suspended solids (TVSS) in each column by adding the values found in each layer. The sludge volume index (SVI) was calculated as ml g^{-1} VSS from the 30 minutes decanted volume and the VSS concentration, because variable presence of fine grains of mineral suspended solids issuing from fillings prevented from comparing TSS results from one column to another.

Flow characterization

In complex porous media filtration, most of studies concern small granulometries and fluid velocities. In this case, flow is dominated by laminar conditions and Darcy's law is valid. When speed and size increase, and as soon Re > 1, the inertial effects are not negligible compared to viscosity forces, and Darcy's law is no more valid [18-20].

Bear [21], reporting earlier experiments, notes that for complex porous media three flow regimes may be identified according to the local Re: as still as Re < 2, Darcy's law is valid, but as Re increases, stream lines start to shift and fixed eddies begin to appear in the diverging areas of the models, inducing first deviation from Darcy's law. Eddies become larger as Re increases, and at Re = 75 turbulence appears and start to spread out, covering the entire domain when Re reaches 180. Recent computer simulation of flow through a regularly rhomboedric packing [22], confirmed that for Re values of 10 and 50 Darcy's law was not valid, with abrupt changes in flow direction, preferred flow paths and inertial cores in the center of some pores.

In porous media, Re is defined by equation (1) [18]:

$$Re = \frac{du\rho}{\upsilon}, \tag{1}$$

where d is the average pore diameter (m), ρ the fluid density (Kg m^{-3}), υ the dynamic fluid viscosity (Kg $m^{-1}s^{-1}$) and u the average flow velocity (m s^{-1}).

The average flow velocity was evaluated in each column as the average downwards speed of the flushes and the values of Re calculated using eq. (1) and a mean measured pore diameter of 1 mm in the zeolites and 0.3 mm in the sand.

On another hand, the usefulness of dimensional analysis to describe the hydrodynamic conditions that prevail in aerated reactors was early pointed out. In this approach, the aeration intensity I (superficial velocity in m s^{-1}) can be linked with the Re through equation (2) [23]:

$$Re = \frac{Id_b}{\nu} \qquad (2)$$

where 'ν' is the kinematic fluid viscosity ($m^2\ s^{-1}$) and d_b the bubble diameter (m).

As only large differences in Re numbers are significative, this number can be used to compare flow regimes occurring in different systems where fluid properties are roughly conserved.

Results and Discussion

During the percolating filters follow-up, ASG were first noticed in the filters effluents around weeks 70 – 80. Two campaigns of microscopic examinations were thereafter conducted on the columns effluents (Table 3).

Typical regular-shaped spherical dense granules were found in the effluents of columns C3 and C4 (4-1 and 4-2), along with a typical aerobic microfauna (*Tardigrata, Rotifera, Thecamoeba* and *Ciliated Protozoa*), but no ASG were found in the effluents of C2 and C5 and very few in the effluents of C1.

At the end of the experimental period, the six columns were dismantled and fillings fractionated in 10 cm layers as a compromise between precision and volume, except the interface sample, which corresponded to the 2-3 cm layer of ASG accumulation area between the two granulometries for dual-layer filters. Layers were identified as A, B, C, D, Interface and E from the top to the bottom of columns respectively and the biofilms extracted.

ASG were observed along with smaller biofilms and typical aerobic microfauna in all columns except the sand column. Their abundance and size varied depending of the layer observed and of the column, as did the other parameters summarized in Table 4.

Figure 2a and Figure 2b show granules developed in the columns.

ASG found here pertained to the smallest range reported by other workers [3]. They formed up to 50% of the observed biofilms volume for the highly loaded filters (C2, C3, C4-1 and C5), especially at or above the interfacial area (layers C and D). Although also present in upper layers A and B and lower layers E, they were smaller and far less abundant.

Biofilm accumulation in the sand filter C1 took place as usual in the upper layers, but ASG were not noticeable at any place, and their abundance in the effluent stayed two orders of magnitude less than in the effluent of filters C3 and C4. This few ASG may have developed in the draining layer of gravel at the bottom of the filter.

The mean of biofilms SVI for all the values obtained when ASG were present was low (58 ml g^{-1}) and typical of values of 50 to 80 ml g^{-1} given for SBR ASG [24]. The mean when they were absent, 232 ml g^{-1}, was typical of poorly decanting activated sludge biofilms.

The total quantity of biofilms and ASG as VSS reached high values in the zeolitic filters, with a mean of 38 g VSS for C2, C3 and C4-1, of the same order that one's found in SBR [24]. This was far above the values for the sand filter (13 g) and the low-loaded zeolitic filter C4-2 (18 g), but far less than for the C5 filter (145 g).

Biofilms occupied an important part of the porosity of the clean media, increasing from the upper layers to the interfacial and lower

column		C1	C2	C3	C4-1	C4-2	C5
filling		sand	chabazite	clino 1	clino 2	clino 2	clino 2
first layer (top)	height (cm)	70	40				
	granulometry (mm)		2-5	2-5	2-5		
	d10 (mm)	0.16	1.5	1.75	1		
	Uc	7.5	1.7	1.4	2.1		
second layer (bottom)	height (cm)	-	15				
	granulometry (mm)		1-2	0.5-2	1-2		0.3-0.7
	d10 (mm)		0.5	0.66	0.7		0.38
	Uc		1.4	1.4	1.4		1.8

Table 1: Granulometrics characteristics of the filings of the experimental columns.

Columns	Reference surface (m^2 per housing)	Surfacic load (cm day^{-1})	Number of flushes per day	flushes timing (24 hours basis)	Water height per flush (cm)	Volume of flushes (L)
C1	25	2,4	4	09; 11:30; 19; 20	0.6	0.1
C2; C3; C4-1; C5	5	12	7	08; 09; 11:30; 19; 20; 21; 22	1.7	0.3
C4-2	10	6	7		0.9	0.15

Table 2: Hydraulic load of the columns.

Period (weeks nb.)	Spherical Granules:	Column					
		C1	C2	C3	C 4-1	C 4-2	C5
104 - 107	abundance 10^4 /l	0.01	-	1	1.5	1	-
	diameter, millimeters	0.25	-	0.2-0.25	0.25-0.5	0.15-0.2	-
117 - 119	abundance 10^4 /l	-	-	1	1.2	1	-
	diameter, millimeters	-	-	0.05-0.2	0.2	0.1-0.2	-

Table 3: Distribution of the ASG abundances and sizes in the columns effluents.

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Parameter	Layer	C1	C2	C3	C4-1	C4-2	C5
Abundance	A	-	-	+	+	-	+
	B	-	+	+	-	-	+
	C	-	++	+++	+	+	+
	D	-	++	++	+++	++	+++
	Interface	-	+++	+	+	+	++
	E	-	+	-	+	-	-
Diameter [milimeters]	A	-	-	0.15	0.15	0.1	0.05-0.1
	B	-	0.3-0.4	0.2	-	-	0.2-0.5
	C	-	0.2-0.3-	0.15-0.2	0.1-0.2	0.15-0.2	0.25
	D	-	0.2-0.3	0.2-0.3	0.15-0.2	0.2-0.3	0.2-0.25
	Interface	-	0.2-0.3	0.25	0.25-0.3	0.2-0.3	0.2-0.25
	E	-	0.2	-	0.1-0.2	-	-
VSS (g L^{-1} of filling)	A	5.01	0.16	1.29	1.28	2.52	4.73
	B	1.66	3.70	1.69	1.44	3.14	10.89
	C	0.38	7.61	1.77	2.99	4.49	18.96
	D	0.50	7.70	3.91	3.92	4.70	37.74
	Interface	1.24	12.58	7.05	0.43	5.98	21.74
	E	0.56	4.45	13.34	2.35	7.18	18.10
TVSS (g)	Column	12.73	41.09	36.18	18.17	35.74	145.41
SVI (ml g^{-1})	A	47	449	56	57	50	71
	B	77	39	43	51	41	47
	C	339	41	82	115	36	45
	D	252	52	37	84	46	37
	Interface	73	35	39	171	58	51
	E	259	59	38	31	13	63
Fraction of initial porosity occupied by biofilms (%)	A	72	18	15	15	27	23
	B	39	35	15	15	27	38
	C	39	75	29	72	34	68
	D	39	98	29	68	45	95
	Interface	28	100	55	15	93	61
	E	44	95	100	20	25	33

Table 4: Spatial distribution and some characteristics of ASG inside the filters (abundance: -: < 10^5 granules l^{-1}, +: < 10% of total agglomerates, ++: < 20% and +++: > 20% = up to 10^6 granules l^{-1}).

layers, especially for C2 and C5 filters where it reached the whole porosity.

Filters C2 and C5 experienced some clogging, with an increasing height of stagnant water (saturated zone) above the interfacial layer from 2 or 3 cm on week 84, to 30 cm on week 110, but the water still percolated slowly through the ASG accumulation and no overflow occurred at the end of the experiment. Water directly sampled in this saturated layer for filter C2 during weeks 99 and 108 showed numerous 0.2-0.3 mm diameter ASG ($1.2*10^6$ l^{-1} of sample, Figure 3), while ASG where absent in its effluent. So, physical retention and accumulation of ASG at the interfacial clogging zone between the two granulometric layers affected filters C2 and C5 only. This phenomenon was correlated to the slightly smaller granulometry of their second layer (cf Table 1). Although statistical evaluation was not possible, ASG found in the effluents of C3 and C4 were slightly smaller than those found inside their upper layers, indicating a partial straining at the interfacial area (Figure 3).

The values of Re calculated applying equation (1) to our experimental conditions are given in Table 5. Excepted for the sand column, the values obtained lay strictly within the range previously defined for fixed eddies conditions (between 2 and 75), above laminar flow and under turbulent flow.

The main similarity that can be found between the operational cycles of SBR and DLPF concerns the Re values. In conventional activated sludge systems, high-speed recirculating pumps drastically increase the mean Re encountered by suspended biofilms, whereas SBR reactors have no recirculating pumps and their suspended biofilms undergo much lower Re values. Values of Re ranging from 10 to 60 were found in full-scale bubble-aerated activated sludge tanks [25]. Values of Re can be calculated applying equation (2) to the results of previous studies on ASG in SBR (Table 6). Even if the bubble diameter used for this comparison is an assumption, because the authors gave only an indication ("small bubbles"), this diameter may reasonably assumed around 1 mm and no more than 2. In any case, the Re values obtained stay within the same range.

ASG formation in our DLPFs was so considered due to the slow formation of small ASG by mature aerobic low-adherent biofilms, under eddy-flow conditions in the upper layers of large granulometry, low-heterogeneity zeolitics fillings. This hypothesis is consistent with the mechanism proposed to explain ASG formation in plastic-packed satured-flow biofilters [26].

The partial retention of the largest fraction of ASG in the larger pores gave way to an accumulation of suspended biomass in large excess compared to fixed biofilms. Depending upon the relative granulometry of the second layer and the size of ASG, ASG may undergo either physical straining or accumulation at the interfacial layer, leading to long-term clogging, either percolation through the lower layer and evacuation with the effluent.

Further modeling of the performances of similar percolating filters

Column	C1	C2	C3	C4-1	C4-2	C5
Percolating velocity (cm.s^{-1})	0.03	2.1	1.38	1.78	1.78	1.78
Reynolds number	0.21	59	39	51	51	51
Mean ASG size, (mm)	--	0.3	0.25	0.2	0.2	0.2

Table 5: Values of Re number inside the DLPFs:

Author and substrate	Superficial velocity (cm s^{-1})	ASG diameter (mm)	Re
Tay et al. 2001a (acetate)	1.2 (min)	0.37	12
id.	2.4 (opt)	0.35	24
id.	3.6 (max)	0.3	36
Tay et al.2001b (glucose)	2.5 (opt)	2.4	25
Tay et al.2001b (acetate)	2.5 (opt)	1.1	25
Beun et al. 2002 (acetate)	2.2	2.5	22
Chen et al. 2008 (acetate)	3.2	0.3	32
id.	2.4	0.6	24
Hailei et al. 2006 (papermaking wastewater)	4	1	40
id.	3	1.4	30
id.	2 (min)	1.8	20

Table 6: Re values calculated from SBR studies assuming 1 mm bubbles diameter, and corresponding obtained ASG diameter.

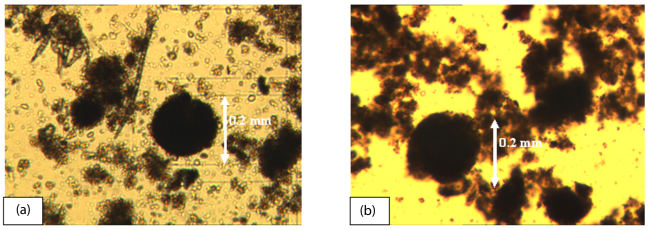

Figure 2: (a) Granules formed in the layer D of column C3; (b) Granules formed in the layer D of column C5.

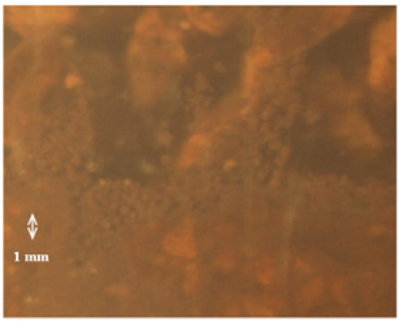

Figure 3: Aerobic granules accumulation at the interfacial area between the two granulometries, column C2, week 91.

cannot thus rely solely upon the current hypothesis of fixed biofilm accumulation on the media superficy [27].

Conclusion

Small typical aerobic spherical granules (ASG) have been found during the long-term follow-up of dual-layer percolating filters (DLPFs). Their development is correlated to the prevailing non laminar flow conditions during the water flushes specific of intermittent filtration, characterized in our experiments by Reynolds numbers between 40 and 60. These conditions can be compared to those prevailing in well-studied ASG-forming SBR. ASG growth appears in DLPFs as an active biomass retention capacity alternative or complementary to fixed biofilm growth that may influence strongly their working capacities (performances and clogging). This must be considered to explain or modelize hydraulical behavior as well as degradative capacities of such filters.

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