Aerobic granulation in a sequencing batch reactor (SBR) for industrial wastewater treatment

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Abstract Aerobic granulation seems to be an a attractive process for COD removal from industrial wastewater, characterised by a high content of soluble organic compounds. In order to evaluate the practical aspects of the process, comparative experimental tests are performed on synthetic and on industrial wastewater, originating from pharmaceutical industry. Two pilot plants are operated as sequencing batch bubble columns. Focus was put on the feasibility of the process for high COD removal and on its operational procedure. For both wastewaters, a rapid formation of aerobic granules is observed along with a high COD removal rate. Granule characteristics are quite similar with respect to the two types of wastewater. It seems that filamentous bacteria are part of the granule structure and that phosphorus precipitation can play an important role in granule formation. For both wastewaters similar removal performances for dissolved biodegradable COD are observed ($>$ 95%). However, a relatively high concentration of suspended solids in the outlet deteriorates the performance with regard to total COD removal. Biomass detachment seems to play a non-negligible role in the current set-up. After a stable operational phase the variation of the pharmaceutical wastewater caused a destabilisation and loss of the granules, despite the control for balanced nutrient supply. The first results with real industrial wastewater demonstrate the feasibility of this innovative process. However, special attention has to be paid to the critical aspects such as granule stability as well as the economic competitiveness, which both will need further investigation and evaluation.

Keywords Aerobic granules; carbon removal; SBR; industrial wastewater

Introduction

Granulation is the process in which biomass forms discrete, well defined, smooth and dense pellets, without any application of support material. This phenomenon is well known under anaerobic conditions. Similar to anaerobic applications, aerobic granulation is characterised by high settling velocity of the granulated biomass leading to very good solid/liquid separation and high biomass accumulation in the reactor. Hence, important volumetric conversation rates and compact installations can be the typical application issues of this technology. It is however less investigated than anaerobic granulation with regard to the influencing parameters for granule formation as well as its applicability to full scale wastewater treatment. Literature reports mainly on studies with synthetic wastewater (e.g. Beun *et al.*, 1999; Tay *et al.*, 2003; Tsuneda *et al.*, 2004). Research on "real" wastewater was adressed by Morgenroth *et al.*, (1997), applying molasses, and Schwarzenbeck *et al.* (2004), utilising a mixture of barley dust and tap water. Without questioning the importance of fundamental and basic research based on synthetic wastewater, it should be stressed that applying industrial/real wastewater is equally important to identify the potential of the process of aerobic granulation and possible problematic conditions.

The aim of this study is to evaluate the practicability of the aerobic granulation process for COD removal from industrial wastewater. For comparative purposes experimental tests are performed on synthetic and industrial wastewater. Applying real industrial wastewater is essential in order to screen the value of the aerobic granules process. Initial focus is put on removal of organic pollutants, applicable loading, operational procedures, parameters necessary for formation and granule stability.

Methods

Reactor set-up and operation

Two independent pilot plants are operated as Sequencing Batch Bubble Columns (SBBC). These reactors consist of cylindrical columns, with a diameter of 200 mm and a total high of 1.8 m. The working volume of the system is 40 L. The initial phase definition of the SBR cycle is as follows: inlet phase (4 minutes), aeration phase (3 hours), settling phase (3 minutes) and discharge phase (2 minutes). A volume exchange rate of 60% is applied, corresponding to a hydraulic retention time of 5 hours. Both columns are equipped with coarse bubble aeration systems. High shear stress, considered as another parameter important for the formation of aerobic granules (Tay *et al.*, 2001), is provided by elevated gas flow velocity (0.74 cm/s).

The reactors are maintained at room temperature (20°C ± 2°C). No pH control is applied, resulting in pH values between 7.5 and 8.0.

The pilot plant running on with synthetic wastewater is seeded with conventional activated sludge. The reactor fed with industrial wastewater, however, is seeded with biomass previously fed with acetate. Allowing granules formation, a low organic load (2 kgCOD/m^3.d) is applied at the beginning of the seeding period in order to allow for an alternation of feast and famine periods. Once granules are formed, the organic load is increased.

Media description

Synthetic wastewater composition is based on literature (Beun *et al.*, 1999), exchanging ethanol with acetate as the main carbon source. Synthetic wastewater is prepared as a concentrated solution and diluted with tap water during the inlet/feed phase.

The industrial effluent originates from the pharmaceutical industry and exhibits a biodegradability of 75–80% (Figure 1). Organic matter is composed of acetic acid, ethanol, methanol and organic products from pharmaceutical synthesis. To avoid degradation of organic compounds during the storage period, the wastewater is kept in a refrigerated tank (4°C). Five different charges of wastewater were used during the experimental period. As a consequence a certain variability, especially in nutrient content, is observed. When needed, phosphorus and ammonium are added to the wastewater. In order to apply similar operational parameters as with synthetic wastewater, e.g. HRT and organic load, the industrial wastewater was also diluted with tap water during the inlet/feed phase. Average pollutant concentrations for both wastewater are presented in Table 1.

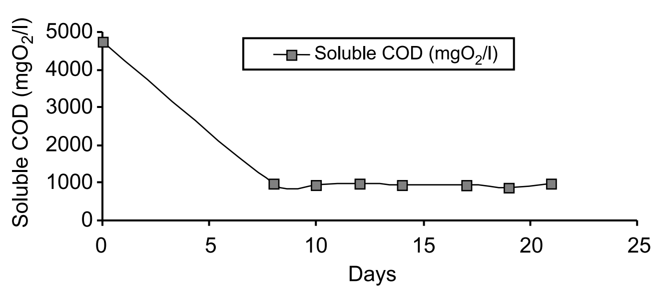

Figure 1 Representative evolution of dissolved COD during an aeration test

Table 1 Average pollutant concentrations of concentrated wastewater

Wastewater	COD_{Total} mg/l	$COD_{Dissolved}$ %	NH_4-N mg/l	PO_4-P mg/l	SS mg/l	pH	Dilution with tapwater
Synthetic	4500	100%	150–225	200–400	0	7	1:10 to 1:3
Industrial	4000–5000	90%	20–300	<5	150–300	4–10	1:10 to 1:5

Analytical procedures

Monitoring of the reactors includes measurement of: suspended solids concentrations (reactor and outlet), biomass ash content, biomass bed volume, granule density and granules settling velocity. NH_4-N, NO_3-N, NO_2-N, PO_4-P, total and filtered COD are frequently followed during a whole cycle by spetrophotometry (Dr Lange).

Results and discussion

This section summarises the results for both types of wastewater, indicating similarities as well as specific differences with respect to granule formation and characteristics as well as their performance concerning COD removal. Results obtained from the industrial wastewater that are discussed in the first sections are referring to the periods of well functioning aerobic granulation. The experimental period with the industrial wastewater exhibited specific problems probably due to the variation of the effluent. A temporary loss of granules with subsequent recovery has been observed. Consequently a sub-section is dedicated especially to the discussion of the observed phenomena during the treatment of the pharmaceutical wastewater.

Granule formation and characteristics

For both types of wastewater, a rapid formation of granules (2 to 4 weeks) was observed along with a high COD removal rate.

According to visual observation, filamentous bacteria are part of the granule structure (Figure 2). Furthermore, microscopic observation seems to indicate that phosphorus precipitation could play an important role in granule formation (Figure 3). Phosphorus precipitation in the reactor could initiate granule formation by providing a first carrier or seeding point for the bacteria to grow on. It can be shown by simple calculation that chemical precipitation is taking place. Hence, it might be possible that the precipitants serve as seeding points for granules formation under the operation conditions applied.

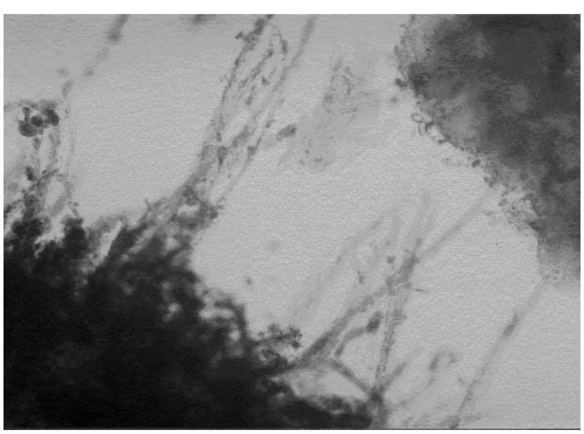

Figure 2 Microscopic observation of granules surface

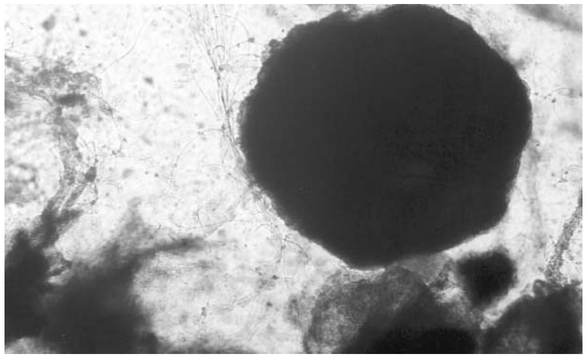

Figure 3 Microscopic observation of granules in formation

This hypothesis seems to be supported by evaluating the percentage of volatile suspended solids of the of the granular sludge (Table 2).

The high proportion of phosphorus in the synthetic wastewater (4 to 8 mg P/100 mg COD) compared to the one of the industrial wastewater (1 mg P/100 mg COD), results in more dense granules with a higher ash content (50% compared to 70 to 80% with industrial wastewater) and with a better settling velocity. Granules characteristics obtained from both types of wastewater are presented in Table 2. Their appearance is similar: they are smooth and dense, and have a very clear outline. The granules obtained from industrial wastewater, however, exhibit a lower density 45 g/l (compared to 50–100 g/l from synthetic wastewater) and hence do have a lower settling velocity. The values determined for the granules from synthetic wastewater are well within the range cited in the literature: Beun *et al.* (2000) stated for the granules formed in a sequential airlift reactor a density of 40 gVSS/l; and Etterer and Wilderer (2001) determined in their study 72 m/h as a mean settling velocity.

Literature suggests that an alternation of feast and famine periods is necessary for granule stability (see also Figure 4). Intermittent feeding according to Beun *et al.* (2002), induces periods of high load with deep penetration of substrate in granules and leads to increasing biomass density and stability. More recently, McSwain *et al.* (2004) demonstrated that long anaerobic fill phase (90 min instead of few minutes), leading to intermittent feeding, affects the selection and growth of floc-forming and filamentous organisms and is necessary for the formation of dense and compact granules. The importance of the famine period was experimentally tested by reducing the famine period during the aeration phase virtually down to zero. A complete loss of granules from the reactor was observed once the time allocated to the famine period was below 30 minutes (results not shown). Re-establishing an adequate famine period resulted in fast recovery of granule formation.

Granule efficiency

Results with synthetic wastewater. Once granules have been formed (Figure 5), high organic loads (7–8 kg COD/m^3.d) were successfully tested with synthetic wastewater. Table 3 displays results for two different levels of applied organic load. A very good

Table 2 Granule characteristics the two types of wastewater

	Synthetic wastewater	Industrial wastewater
Granules density	> 100 gSS/l	50 gSS/l
Granules settling velocity	100 m/h	50 m/h
%VSS	50%	70–80%

Table 3 Results with synthetic wastewater (mean values for several weeks of operation)

Applied load (kgCOD/m^3.d)		Inlet WW (mg/l)			Outlet WW (mg/l)			Eliminated load (kgCOD/m^3.d)	
COD_T	COD_D	SS	COD_T	COD_D	SS	COD_T	COD_D	COD_T	COD_D
2.7	2.7	0	540	540	260	228	23	1.75	2.5
7.1	7.1	0	1380	1380	350	393	57	5.4	6.4

dissolved COD removal rate was reached (95%) as acetate is an easily biodegradable organic compound. However, a relatively high concentration of suspended solids in the outlet causes a lower performance of total COD removal. Biomass detachment seems to play a non negligible role in the current set-up. This phenomenon is not stated explicitly in the literature, yet calculations with the corresponding values from literature do underline the same problem. This aspect has to be solved as it represents a major drawback of the process so far. The phenomenon is not fully investigated, but a recent study achieved suspended solids concentrations of around 20 mg/L in the outlet (Van Loosdrecht, personal communication). This demonstrated the general possibility to overcome this crucial aspect.

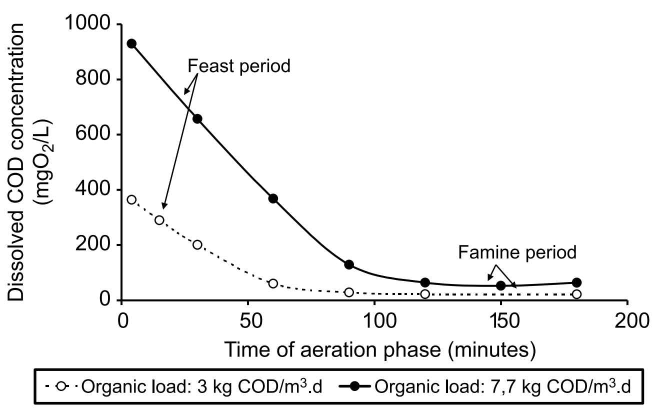

Figure 4 COD removal during a cycle with synthetic wastewater

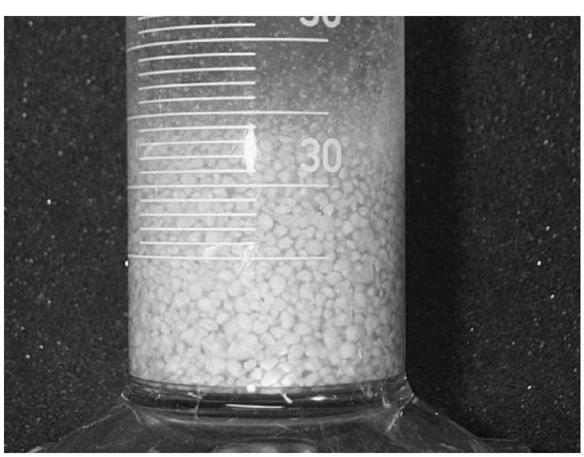

Figure 5 Granules obtained with synthetic wastewater (organic load = 2.5 kgCOD/m^3.d

Figure 4 presents the dissolved COD consumption during the aeration phase for different applied organic loads. Dissolved COD is rapidly removed in the reactor and a long famine period can be observed: 50 to 70% of the aeration phase is a famine period. This might not be the optimised value, however as stated above, an important reduction in the famine period induces the risk of rapid granule destruction.

Results with industrial wastewater. Once granulation has begun with industrial wastewater (Figure 7), the applied load was increased from 3 to 5.5 kg $COD/m^3.d$. Table 4 presents a summary of the results obtained with industrial wastewater. The dissolved COD concentration in the outlet corresponds to the refractory or non biodegradable part of the effluent (20% of influent COD). The elevated total COD and SS concentrations in the outlet are mainly due to the suspended solids in the inlet wastewater that are not removed in the reactor and to biomass detachment from the granules surface. Figure 6 shows the dissolved COD consumption during the aeration phase. This illustrates that this process treats soluble COD, but particulate pollutants should be removed via appropriate pre-treatment. Furthermore, it emphasises that the operation of the reactors still have to be optimised in order to reduce biomass detachment to a minimum. Recently, Schwarzenbeck *et al.* (2004) have experimentally determined that particles up-take can occur on the granules surface for a certain size of particles. Further studies are needed in this direction in order to determine the essential operational or design parameters for optimised suspended solids retention within the process.

Granule stability when treating industrial wastewater

The pilot plant fed with industrial wastewater exhibited a temporary disruption of the aerobic granule process during the testing phase of this study. The experimental period can be divided into 3 distinct segments, as illustrated in Figure 8.

During the 1st phase, an organic load of 3 kg $COD/m^3.d$ is applied during start-up in order to favour granule formation. A rapid granule formation could be observed

Table 4 Results with industrial wastewater (mean values for several days of operation)

Applied load ($kgCOD/m^3.d$)		Inlet WW (mg/l)			Outlet WW (mg/l)			Eliminated load ($kgCOD/m^3.d$)	
COD_T	COD_D	SS	COD_T	COD_D	SS	COD_T	COD_D	COD_T	COD_D
3.2	2.9	76	640	570	108	246	97	2.1	2.4
5.5	5.0	100	1085	985	194	505	216	3.1	3.9

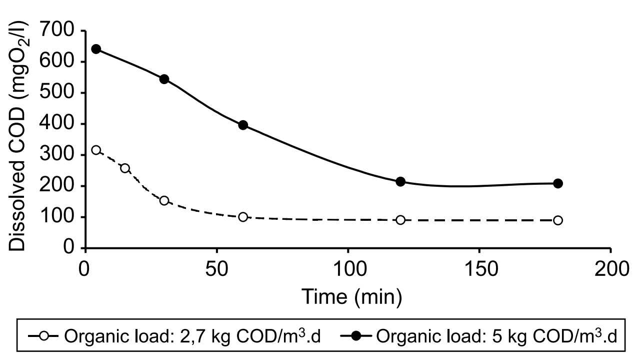

Figure 6 COD removal during a cycle with industrial wastewater

Figure 7 Granules from industrial wastewater (Load = 3 kgCOD/m^3.d)

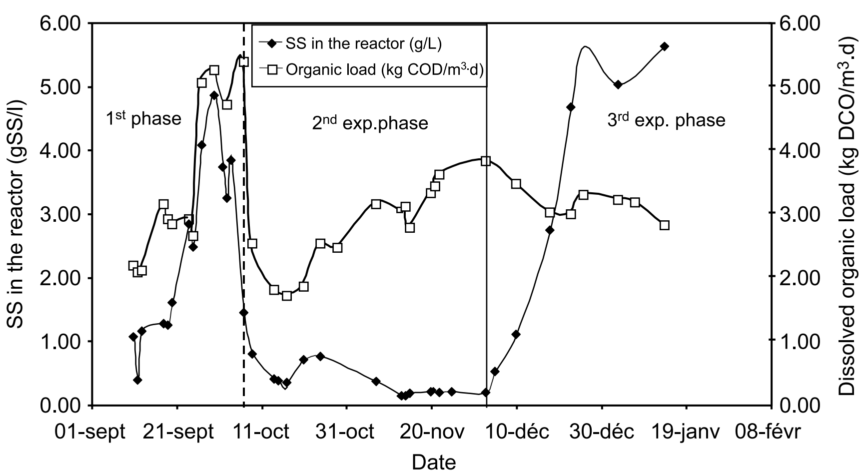

Figure 8 Evolution of suspended solids and organic load – testing industrial wastewater

(1–2 weeks), illustrated by the evolution of the suspended solids concentration in the reactor (Figure 8).

As a good dissolved COD removal rate was rapidly obtained, the applied load was increased (to 5.5 kg COD/m^3.d.). This induced a further increase in the SS concentration, being related to the increased granules in the reactor. However, a rapid disappearance of granules is observed after the 7 October, which coincides with applying a new charge of the industrial wastewater. Development of filamentous bacteria was observed on the granule surfaces. Granules were rapidly transformed in big pellets that were not settling any more but were floating, inducing a rapid wash out of all biomass.

The 2nd phase is characterised by the virtual absence of granules or even floc forming biomass in the reactor and very low SS values. As previously explained, after the change of wastewater, granules rapidly disappeared in the reactor. The important development of filamentous bacteria on the granule surface could be explained by the complete lack of phosphorus in this charge of the wastewater. Hence, external phosphate addition was started at the same ratio, as found in the first charge 1 mgP/500 mgCOD. In parallel the organic load was decreased to 2–3kg COD/m^3.d to help granule formation. As can be observed on Figure 8, until the beginning of December nothing happened in the reactor. No biomass was retained in the reactor, no granules were formed. However, a removal rate of 60–70% for soluble COD was observed, probably due to the biofilm formed on the reactor walls as well as pin floc formation in suspension.

The 3rd phase starts with the increase in external phosphorus addition (ratio of 1 mgP/100 mgCOD) from the beginning of December. At once a rapid biomass growth is noted in the reactor (Figure 8). Granules are rapidly formed and the biofilm detaches from the pilot wall and a removal rate of 80% for filtered COD is reached.

The results obtained during phase 3 are very similar to the ones noted during the first phase of experiments with the same organic load. Virtually all soluble, biodegradable COD is eliminated. Hence the residual COD_f in the outlet corresponds to the 20% of refractory dissolved COD.

This incident illustrates the risk with regard to granule stability. It seems that phosphorus addition has helped to overcome the problem and hence might underline the importance of precipitation products for granule formation. However, it is believed that P-precipitation is not the only important parameter and that other parameters probably play an equally important role. As detailed analysis of the wastewater is missing, nothing can be said about a possible component that might trigger granule destabilisation. The gas flow velocity applied in this study (0.74 cm/s) is below the values (1.2; 2.4; 3.6 cm/s) tested by Tay *et al.* (2003) for supplying adequate shear stress for dense and smooth granule formation. This could indicate that during the study presented here, maybe the granules were not supported enough by adequate shear stress.

The high variability of industrial wastewater seems to be a potential risk concerning granules stability. Their robustness with respect to dynamic conditions as well as granule evolution with time must be further investigated. From the literature it appears that selection for slow growing micro-organisms influences positively stability of granules (De Kreuk and van Loosdrecht, 2004). Selection of slow growing micro-organisms can be obtained thanks to prolonged anaerobic feeding period.

It should be stated here, that it is known that the applied wastewater is treated by an activated sludge plant without observing major malfunctioning, as long as sufficient nutrient presence is assured. Hence in order to be competitive the granule stability issue has to be resolved and clearly needs further investigation.

Conclusion

The aerobic granule process represents a very interesting process that could be situated between high loaded activated sludge and anaerobic treatment with regard to COD removal. It is designed for soluble COD removal, calling for adequate pre-treatment or post-treatment to remove detached biomass. It aims at low footprint reactors with a low sludge production. This study focuses preliminarily on COD removal, but literature reveals also its potential for N and P removal (Beun *et al.*, 2001).

For both types of wastewater good dissolved COD removal is observed (95% and 80% at a load of 7–8 kg COD/m^3.d and 5.5 kg COD/m^3.d. for synthetic and industrial wastewater respectively). The lower performance on the pharmaceutical wastewater can be attributed to content of 20% of refractory COD. Hence, the first results with real industrial wastewater demonstrate the feasibility of this innovative process. However, a relatively high concentration of suspended solids in the outlet deteriorates the performance with regard to total COD removal. Biomass detachment seems to play a non negligible role in the current set-up. It emphasis that the operation of the reactors still needs further optimisation in order to reduce biomass detachment to a minimum. In addition, special attention has to be paid to the critical aspects such as granule stability as well as the economic competitiveness, which will both need further investigation and evaluation.

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