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Aerobic granular sludge to treat paper mill effluent: organic matter removal and sludge filterability

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ABSTRACT

Recently, researchers working with aerated bioreactors identified a different microbial organization in the form of aerobic granules. In this configuration, bacteria clump together more tightly than conventional floc, which reduces biofilm formation on the membrane surface of membrane bioreactors and allows higher flux for longer time periods during filtration. This study examined the possible formation of aerobic granules treating paper mill effluent and compared the removal efficiency of organic matter from granular with the conventional flocculent sludge. The filterability of both types of sludge was also compared. Two reactors were operated in parallel with aerobic granular and flocculent sludge. Both systems achieved very high Biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD)-removal efficiencies. Sludge filtration tests showed a higher critical flux for granular sludge with total suspended solids (TSS) equal to 2,700 mg L^{-1}. However, the opposite behavior was observed when the reactors were operated with a TSS value of 4,400 mg L^{-1}.

Keywords: Aerobic granular sludge; Paper mill effluent; Sludge filterability

1. Introduction

Paper mills generate large volumes of effluent rich in organic matter. Depending on the type of paper produced and paper machine characteristics, water consumption can be as high as 100 m^3 per ton of paper produced. The conventional activated sludge (AS) process has been widely used for the treatment of paper mill effluent to achieve a high efficiency in the removal of organic matter. In the conventional AS process, micro-organisms in the aeration tank typically form biological flocs. Flocculation is the result of microbial metabolism and most often occurs under limited food availability or some other type of stress [1].

Recent studies have shown that under certain conditions, the microbial aerobic community can form granules. The morphology of aerobic bio granules is completely different from any other biofilm types, such as that found in AS flocs [2,3]. Aerobic granular sludge can be a remarkable approach to overcome problems related to sludge settling ability in conventional AS [4]. Several recent publications have demonstrated the feasibility to form aerobic granular sludge but using essentially synthetic effluent [4–8]. However, currently, there is little knowledge on the efficiency of granular sludge to remove organic matter from wastewater

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compared to conventional flocculent sludge and there are no publications on the formation and use of aerobic granular sludge to treat paper mill effluent.

In a membrane bioreactor (MBR), the secondary clarifier is replaced by a membrane filtration process that retains suspended solids in the system (concentrate) and allows for the passage of treated effluent (permeate). The main advantage of this system over the conventional AS is the production of a treated effluent free of suspended solids. The major limitation of the MBR process is the occurrence of membrane fouling and consequently a reduction in the permeate flux.

Granular sludge can potentially result in better settling and filterability characteristics because of the greater size of the flocculated sludge. This characteristic coupled to the low compressibility of granular sludge may lead to a significant reduction in MBR membrane fouling and improve permeate fluxes [8]. Currently, the behavior of granular sludge in MBR is still not well understood. Nevertheless, this novel process can become an important tool for the treatment of domestic and industrial wastewater, including paper mill effluent.

2. Objectives

The aim of this study was to assess the technical feasibility of treating paper mill effluent with aerobic granular sludge. The specific objectives are the following: (i) to produce aerobic granular sludge in reactors treating paper mill effluent; (ii) to compare the performance of granular aerobic sludge with conventional flocculated sludge for organic matter removal; and (iii) to assess the filterability of the granular sludge compared to the conventional flocculated sludge.

3. Material and methods

3.1. Effluents and biological sludge

Effluents were collected from an old corrugated cardboard paper mill that produces approximately 5,500 tons per month and has a water consumption of 10 m^3 ton^{-1}. The samples were collected weekly and transported to the Pulp and Paper Laboratory of the Universidade Federal Vicosa and stored at 5˚C. The initial biological sludge was collected from an AS treatment plant of a kraft pulp mill.

Table 1
Sludge characteristics over time

Day	Settling time (min)	Observation
0	20	Flocculent sludge
7	10	Flocculent sludge
10	8	Flocculent sludge
33	6	First granules
38	4	Gradual granule increase
43	2	Most granular sludge
51	1	Completely granular sludge

3.2. Biological treatment

Biological treatment was performed in sequencing batch reactors. Two reactors were operated in parallel: one, a conventional activated sludge and another with aerobic granular activated sludge. Each system had a 6 L aeration tank, and 3 L of treated effluent were removed and replaced with new effluent for each cycle. The reactors were maintained at room temperature. Oxygen was injected in the reactors by air pumps connected to diffusers to maintain the dissolved oxygen (DO) over 2 mg L^{-1}. The cycle was 12 h and the hydraulic retention time was 24 h (3 L was replaced in each cycle and the reactor volume was 6 L). Minutes before completing this 12 h period, the aeration was ceased to allow sedimentation of the biological sludge. The settling period was 1 h in the conventional AS reactor, and the settling time started at 20 min and was gradually decreased to 1 min in the aerobic granular sludge reactor (the sludge characteristics over time are shown in Table 1). DO was maintained above 2 mg L^{-1}, and the effluent pH was between 6.5 and 7.5 before addition of nitrogen and phosphorus to the reactors in the ratio of 100:5:1 for biochemical oxygen demand (BOD_5):N:P. The biomass concentration in the reactors was estimated by the amount of volatile suspended solids (VSS) according to the APHA [9]. Chemical oxygen demand (COD) analysis of the treated influent and effluent solid series and microscopic observation of the biological sludge were performed daily.

3.3. Microscopic observation

Microscopic observations were performed daily on fresh sludge samples using a LEICA model DMLS optical microscope with phase contrast. The granule development and morphological characteristics of the biological sludge were observed during the granulation process. A digital Nikon, model COOLPIX 4500 camera was used to observe photomicrographs of the granules and the morphological characteristics of the biological sludge.

3.4. Effluent characterization

Effluents were characterized before and after treatment daily for the following parameters: COD,

BOD_5, pH, and suspended solids, as recommended by the standard methods for the examination of water and wastewater [9].

3.5. Microfiltration membranes and determination of the critical flux of the biological sludge

In order to obtain information on the capacity of the filterability of the aerobic granular sludge and compare it with the flocculated sludge, critical flux determination was carried out using polymeric membranes. The results could envision the potential applicability of aerobic granular sludge in MBR.

Modules of hollow fiber, submerged microfiltration membranes were used as bench-testing units. These membranes were manufactured and kindly provided by PAM-Selective Membranes, RJ, Brazil .The average membrane pore diameter was 0.2 μm and the membrane surface area was 0.5 m^2. A schematic flow diagram of the system is presented in Fig. 1.

The determination of the critical flux was carried out on the membrane system according to methodology presented by Bacchin et al. [10]. The sludge was transferred from the bioreactors to the membrane system composed of a 5 L storage tank and an injection system of compressed air at the bottom. Suction of the treated effluent was performed using a model CVE-8701- F-ESP series 9035-9 pump, and the pressure was measured by a brand PAM model F-55375 L manometer. At regular intervals, a beaker was used to measure the flux through the filtrate volume for 30 s. Controlling the hydraulic pump rotation and/or opening a needle valve allowed for adjustment of the difference in the suction pressure.

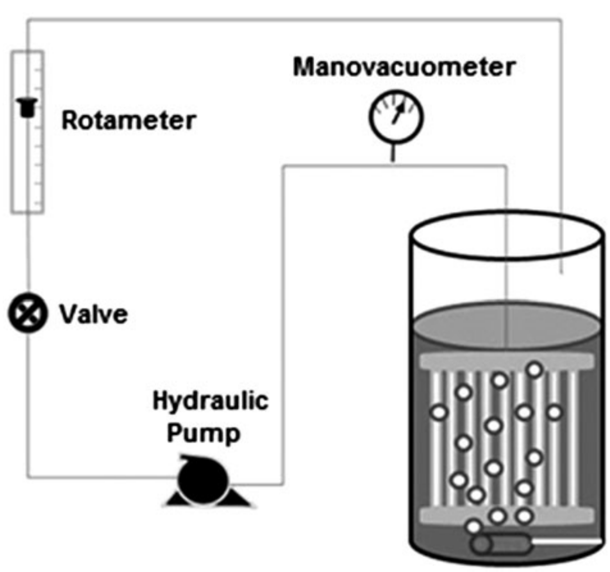

Fig. 1. Schematic flow diagram of the microfiltration system.

Distilled water was used on a clean membrane to assess its resistance before determining the critical flux. Each biological sludge (granular and flocculated) was then added separately to the system, and a set pressure value was used to measure the flux through the membrane during 15 min. Subsequently, the pressure was increased and the flux was measured again during 15 min. The critical flux corresponds to the value below which the system presents variation in the flux, although the pressure is kept constant. Analysis of the total suspended solids (TSS) in the biological sludge samples was performed and the TSS concentration was adjusted by discarding the supernatant from the most diluted sludge. Tests were performed in both systems (flocculent and granular) at the same concentration to minimize the effect of concentrations of solids on the flux. The tests were performed with a TSS of 2,700 mg L^{-1} for the first trial, and the tests were repeated with values of TSS of 4,400 mg L^{-1} for the second trial.

4. Results and discussion

4.1. Aerobic granular sludge formation

Operation of the biological reactor started with the use of biological sludge from an AS sludge treatment plant of a bleached kraft pulp mill. The initial TSS of the sludge was 2,000 mg L^{-1}. An initial settling time of 20 min was used, and consequently, a reduction in the concentration of the TSS was observed during the first days of operation, i.e. there was a significant loss of sludge in the treated effluent.

However, over time, an increase in the TSS concentration was observed. When the TSS in the reactor reached 2,000 mg L^{-1}, the settling time was reduced by half, i.e. 10 min. Again, some sludge loss was observed but when the TSS sludge concentration in the reactor reached around 2,000 mg L^{-1}, the settling time was reduced to 8 min. This procedure was repeated and the settling time was gradually reduced to 6, 4, 2, and 1 min, keeping always the SST concentration close to 2,000 mg L^{-1}. Table 2 presents the number of cycles required to recover the sludge TSS to 2,000 mg L^{-1} for each decrease in phase of the settling time.

Fig. 2 shows photomicrographs of the sludge as the settling time was reduced. The morphological change in the sludge was visible. Flocculated sludge with open flocs was observed initially. Filamentous bacteria were rarely observed in this sludge. As the settling time decreased, there was a predominance of large flocs, culminating with the formation of granules. The number

Table 2
Number of cycles needed to recover a sludge TSS of 2,000 mg L^{-1}

Settling time (min)	Number of cycles
20	–
10	14
8	6
6	46
4	10
2	10
1	16

of filamentous bacteria remained low throughout the process of granule formation.

Fig. 3 shows a photograph of the granules on a Petri plate. The average size of the observed granules, after steady state operation, was approximately 3.5–4.0 mm.

4.2. Removal efficiency of organic matter

Table 3 shows the mean values characterizing the influent and effluent of each system and the average removal efficiency of soluble BOD_5 and COD.

High efficiency in the removal of organic matter (COD and BOD_5) was observed in both systems. The removal efficiency was very similar in both systems, which demonstrated the high removal capacity of the organic matter by the granular sludge.

Because the effluent was collected directly from the industry, there was a significant variation over time in the physical–chemical parameters of the effluent prior to biological treatment. Consequently, there were variations in the treated effluent from both bioreactors that are indicated by standard deviations in the analysis, as presented in Table 3.

However, the pH did not vary significantly between reactors. The average TSS and VSS values of

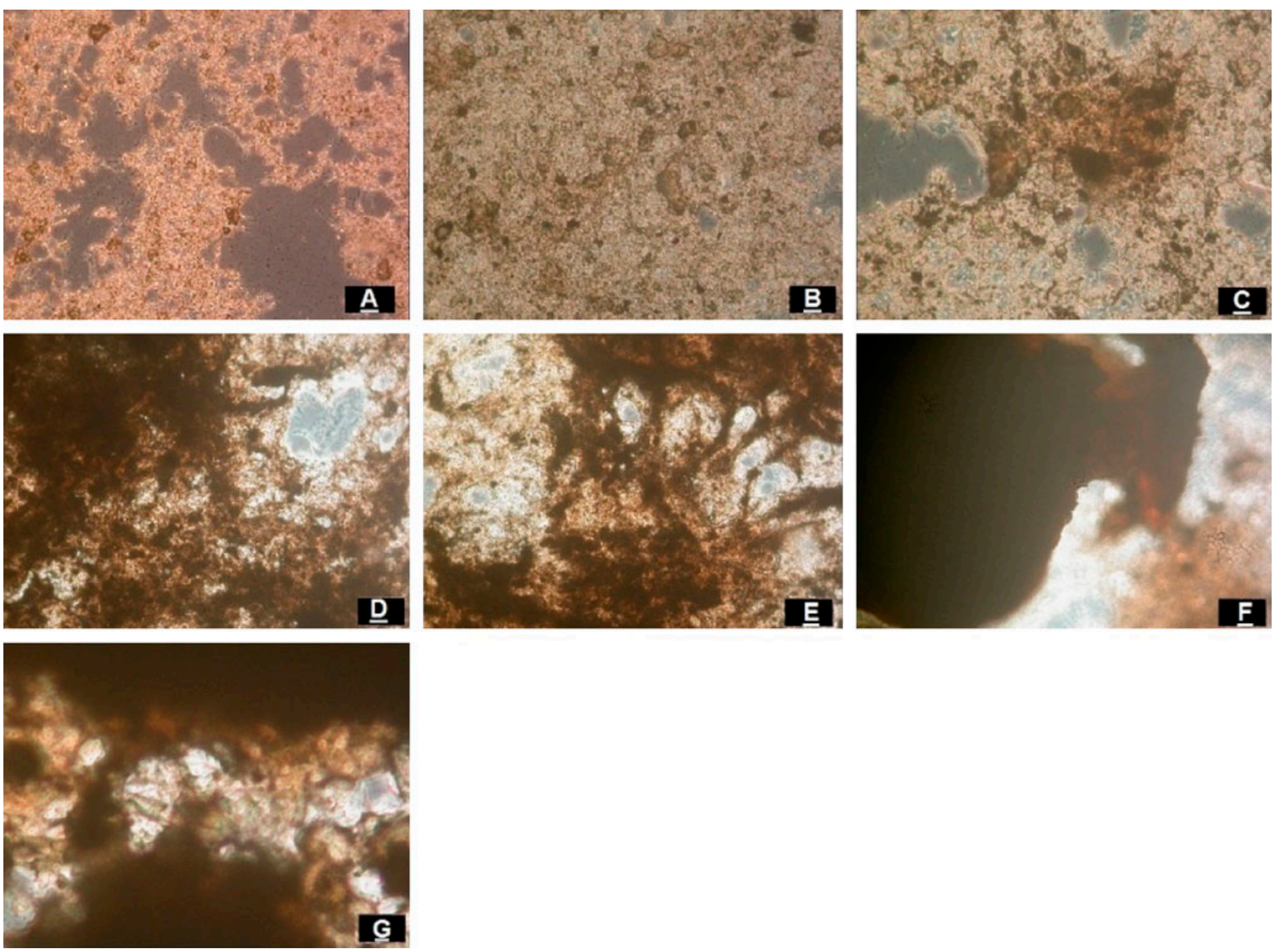

Fig. 2. Photomicrographs of the granular sludge reactor as settling time was reduced. (1)–(3) Flocculated sludge; (4) beginning of granule production; (6) majority of sludge formed by granules; (7) granular sludge.

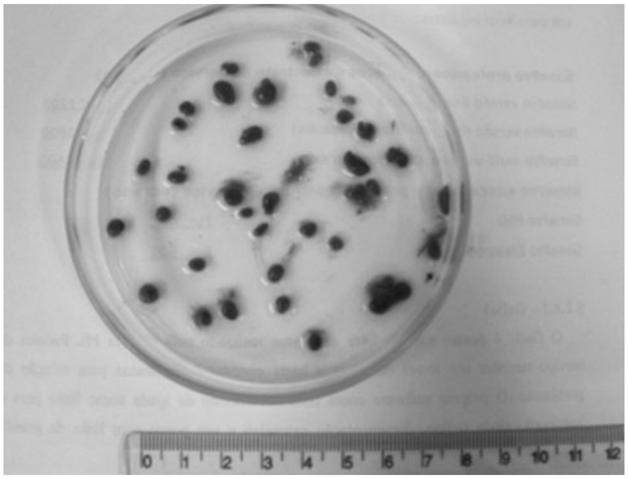

Fig. 3. Aerobic granules on a Petri plate.

the treated effluent in the granular reactor were similar to those of the treated effluent in the conventional AS reactor. Data for the TSS and VSS measurements in the effluent from the granular reactor were obtained only after stabilization of the process with a settling time of 1 min.

4.3. Aerobic granules

The granules have a higher density and, consequently, higher settling velocity because these particles are larger and more compact than the flocs. The settling velocities were 10.8 and 0.18 m h^{-1} for the granules and flocs, respectively.

4.4. Critical flux determination

The critical flux was determined for TSS values of 2,700 and 4,400 mg L^{-1}.

4.4.1. TSS at 2,700 mg L^{-1}

Fig. 4 shows the flux and transmembrane pressure (TMP) values obtained during determination of the critical flux for the aerobic granular and flocculent sludge.

As shown in Fig. 4, filtration with the granular sludge did not present the expected results for critical flux determination at the TSS concentration of 2,700 mg L^{-1}. As observed, there was no major flux reduction during the 15 min of filtration for any of the applied pressure. However, values greater than 0.5 bar could not be obtained with the filtration system using granular sludge at the concentration of 2,700 mg L^{-1}.

In the filterability test of the flocculent sludge shown in Fig. 4, the flux during the filtering test at a TSS concentration of 2,700 mg L^{-1} resulted in a significant reduction when the pressure difference was equal to or greater than 0.4 bar. The critical flux was below a TMP of 0.4 bar and therefore the critical flux was found to be between 100 and 127 L m^{-2} h^{-1}. The average value was equal to 113.5 L m^{-2} h^{-1}.

Table 3
Mean values of parameters analyzed in the reactor influent and effluent

Parameters	Units	Influent	Granular effluent	Conventional effluent
pH	–	6.40 ± 0.70[a]	7.41 ± 0.66[b]	7.46 ± 0.73[c]
Temperature	℃	–	23.7 ± 2.0[b]	23.8 ± 1.9[c]
TSS	mg L^{-1}	566 ± 536[d]	316 ± 306[e]	267 ± 231[e]
VSS	mg L^{-1}	405 ± 391[f]	209 ± 253[e]	185 ± 201[e]
COD_{sol}-Removal efficiency	mg L^{-1} %	780 ± 390[g]	76 ± 44[g]	61 ± 49[i]
			90 ± 7[h]	92 ± 6[h]
BOD_{5sol}-Removal efficiency	mg L^{-1} %	488 ± 268[j]	13 ± 7[j]	12 ± 6[j]
			97 ± 2[j]	97 ± 2[j]

[a]Mean ± standard deviation, n = 169.
[b]Mean ± standard deviation, n = 166.
[c]Mean ± standard deviation, n = 164.
[d]Mean ± standard deviation, n = 75.
[e]Mean ± standard deviation, n = 28.
[f]Mean ± standard deviation, n = 74.
[g]Mean ± standard deviation, n = 59.
[h]Mean ± standard deviation, n = 57.
[i]Mean ± standard deviation, n = 58.
[j]Mean ± standard deviation, n = 23.

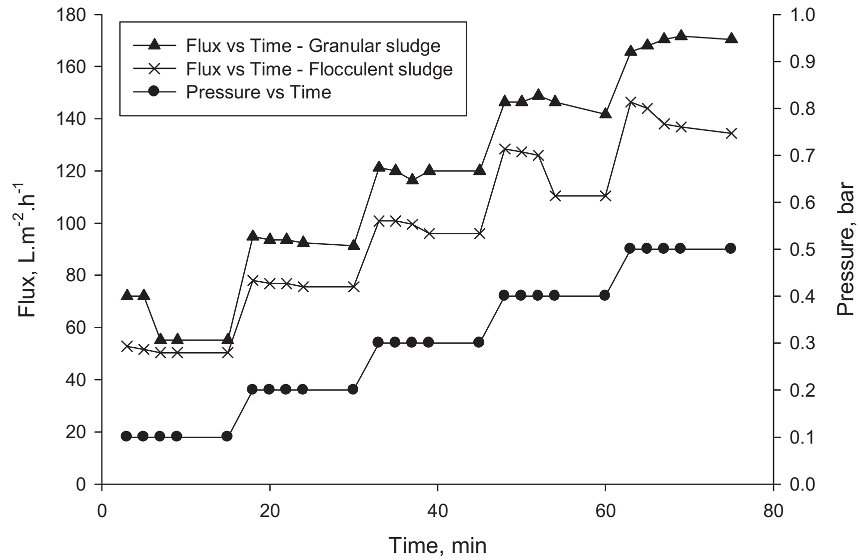

Fig. 4. Flux vs. time for granular and flocculent sludge at different TMP (TSS = 2,700 mg L^{-1}).

High flux values were observed during the filtration of granular sludge compared to flocculent sludge. On average, there was an increase in flux of approximately 17%.

4.4.2. TSS at 4,400 mg L^{-1}

As the critical flux could not be achieved for granular sludge at TSS of 2,700 mg L^{-1}, the filterability test was performed for the sludge at a higher TSS concentration (4,400 mg L^{-1}).

Fig. 5 shows the values of flux and TMP obtained during determination of the critical flux for aerobic granular and flocculent sludge.

Similar to the previous test, the granular sludge at TSS = 4,400 mg L^{-1} did not present any flux reduction at a TMP of 0.7 bar. The critical flux for the flocculent sludge is below a TMP of 0.5 bar, and the critical flux was between 120 and 141 L m^{-2} h^{-1}. The average value was equivalent to 130.8 L m^{-2} h^{-1}.

In contrast to the test with a TSS of 2,700 mg L^{-1}, the observed flux of the aerobic granular sludge was

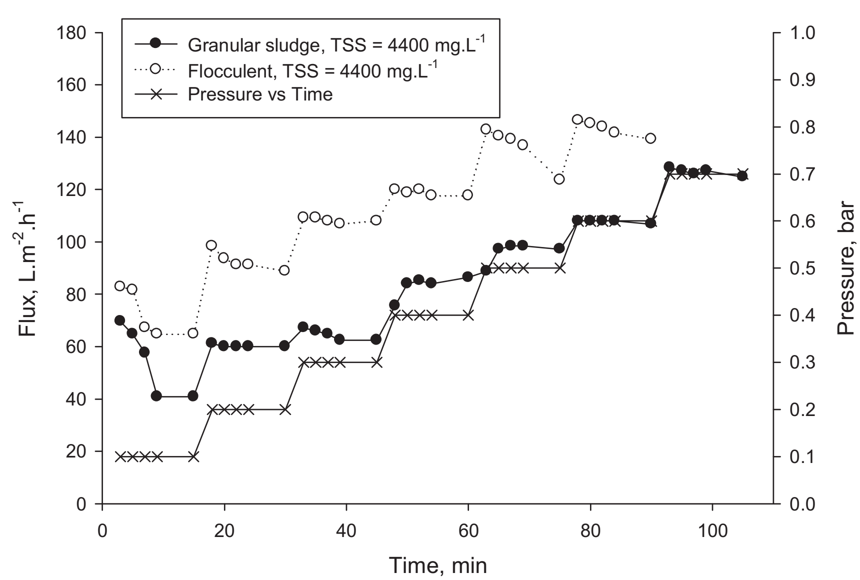

Fig. 5. Flux vs. time for granular and flocculent sludge at different pressures (TSS = 4,400 mg L^{-1}).

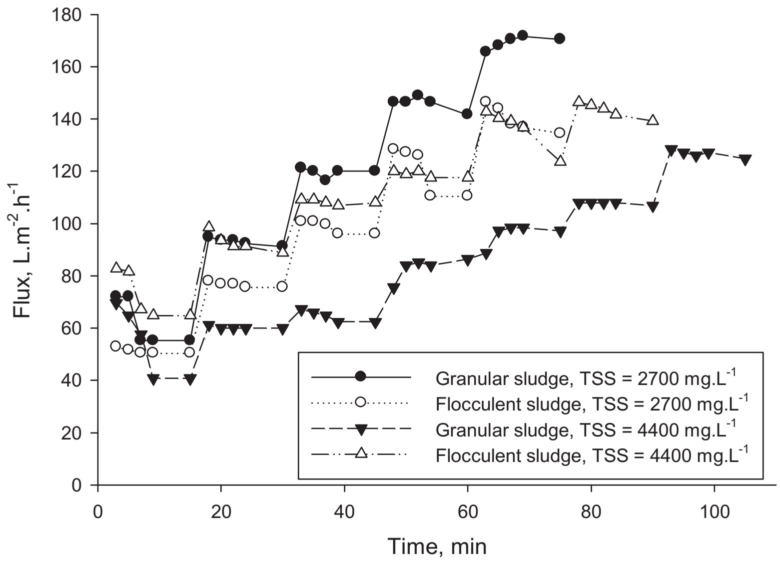

Fig. 6. Flux over time for both granular and flocculent sludge at TSS values of 2,700 and 4,400 mg L^{-1}.

lower than that of the flocculent sludge by approximately 31%. The granular sludge may have suffered structural damages, and dispersed biomass was observed during this test. The granule sludge damages were observed after shear stress occurred during microfiltration. Even after one week of adaptation, the sludge granules never recovered their initial morphology. This result indicated that the granules are not resistant to shear forces and do not recover rapidly from damage.

The granule disruption can also be related to the complex nature of industrial effluent. Lemaire et al. [11] revealed in their study that the granules formed from disruption do not retain their original structure. The influence of industrial wastewater quality and shear forces that damage granules can have important roles in sludge filtration, i.e. sludge granule integrity must be maintained to minimize membrane fouling.

Fig. 6 shows the flux behavior over time for both granular and flocculent sludge at TSS values of 2,700 and 4,400 mg L^{-1}.

As shown in Fig. 6, the flux behavior from the flocculent sludge did not exhibit large variations at either TSS concentration. However, a significant decrease in flux was observed during filtration of the granular sludge. The fluxes were approximately 62% higher for a TSS of 2,700 mg L^{-1} than the fluxes obtained for a TSS of 4,400 mg L^{-1}. As discussed previously in this paper, these results are probably related to granule disruption during the filtration test.

5. Conclusions

Aerobic granular sludge was formed treating paper mill effluent. The performance of COD removal from granular sludge was similar to removal from flocculent sludge, i.e. 90% of COD removal. Granular sludge presented higher microfiltration fluxes than flocculent sludge at a TSS of 2,700 mg L^{-1}. However, this behavior was not confirmed at a TSS of 4,400 mg L^{-1}. This result was most likely due to granule disruption during filtration.

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