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Removal of Nitrogen and Organic Matter in a Radial-Flow Aerobic-Anoxic Immobilized Biomass Reactor Used in the Posttreatment of Anaerobically Treated Effluent

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Abstract This work reports on the removal of organic matter and nitrogen in a radial-flow aerobic-anoxic immobilized biomass (RAIB) reactor fed with domestic sewage pretreated in a horizontal-flow anaerobic immobilized biomass (HAIB) reactor. Polyurethane foam was used as support material for biomass attachment in both reactors. In batch experiments, a first-order kinetic model with residual concentration represented the organic matter removal rate, whereas nitrogen conversion followed a pseudo-first-order reaction in series model, with kinetic constants k_1 (ammonium to nitrite) and k_2 (nitrite to nitrate) of 0.25 and 6.62 h^{-1}, respectively. The RAIB reactor was operated in continuous-flow mode and changes in the airflow rate and hydraulic retention time were found to interfere in the apparent kinetic constants to the nitritation (k_1) and nitratation (k_2). Nitrification and denitrification were achieved in the partially aerated RAIB reactor operating with hydraulic retention times of 3.3 h and 2.7 h in the aerobic and anoxic zones, respectively. Ethanol was added in the anoxic zone of the reactor to promote denitrification. The effluent flow of the RAIB reactor presented a COD of 52 mg l^{-1}, and concentrations of 2 mg $N-NH_4^+ l^{-1}$, 1.24 mg $N-NO_2^- l^{-1}$ and 3.46 mg $N-NO_3^- l^{-1}$.

Keywords Nitrification · Denitrification · Domestic sewage · Anaerobic treatment · Posttreatment

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Introduction

Application of anaerobic reactors for domestic sewage treatment has increased worldwide, mainly in tropical and subtropical countries. Low production of excess sludge, energy saving, and low operational costs are the main reasons for the adoption of such a biological technology. However, according to Chernicharo [1], anaerobic reactors hardly produce effluents that comply with usual discharge standards established by environmental agencies. Therefore, a posttreatment unit is usually required for removal of nutrients and residual organic matter.

The combination of anaerobic and aerobic process can produce effluents that meet environmental standards while consuming less electricity. These combined systems are smaller and produce less sludge than aerobic systems. Moreover, according to Foresti et al. [2], the development of posttreatment units of anaerobic reactors is not only important to improve the effluent quality for environmental protection, but also to achieve the recovery of resources. Several anaerobic–aerobic systems have been proposed for domestic sewage treatment [3–5], most of them combining UASB reactors with activated sludge systems.

Immobilized biomass reactors can be applied as alternative technologies for domestic sewage treatment that combines anaerobic and aerobic processes. Systems with immobilized biomass allow for the use of more compact units operating without recirculation and separation systems. The effective control of cellular retention time, the possibility to achieve high biomass concentration and, consequently, the application of low hydraulic retention times are advantages that stimulate the adoption of immobilized cell technology. However, fixed-bed reactors used for treating sewage lack effective mechanisms for controlling film thickness and may thus contribute to higher mass transfer resistances and to bed clogging.

Recently, Vieira et al. [6] developed a radial-flow aerobic immobilized biomass reactor for posttreatment of domestic sewage previously treated in horizontal-flow anaerobic immobilized biomass reactors. The system performed well in treating domestic sewage, showing good overall removal efficiencies of organic matter (such as COD), SSV, and *N*-ammonium of 90, 94, and 90%, respectively.

The purpose of this work was to evaluate the performance of an aerobic-anoxic radial-flow aerobic immobilized biomass reactor (RAIB) in the removal of organic matter and nitrogen from domestic sewage pretreated in an anaerobic fixed-bed reactor. Kinetic parameters for organic matter removal and for the nitrification process were estimated in batch mode and the RAIB reactor was operated in a continuous mode to evaluate simultaneous nitrification and denitrification, and organic matter removal.

Materials and Methods

The research was carried out in two steps. The first step, consisting of batch experiments, involved estimating kinetic parameters for nitrification and for removal of organic matter. In the second step, the reactor was operated continuously to determine the best operational condition for the nitrification process and to achieve nitrification and denitrification in the reactor by adding an external source of carbon (ethanol).

Reactors

The experimental investigation was carried out in a radial-flow aerobic immobilized biomass (RAIB) reactor (Fig. 1), developed by Vieira et al. [6] and fed with domestic sewage pretreated in a horizontal-flow anaerobic immobilized biomass (HAIB) reactor, as

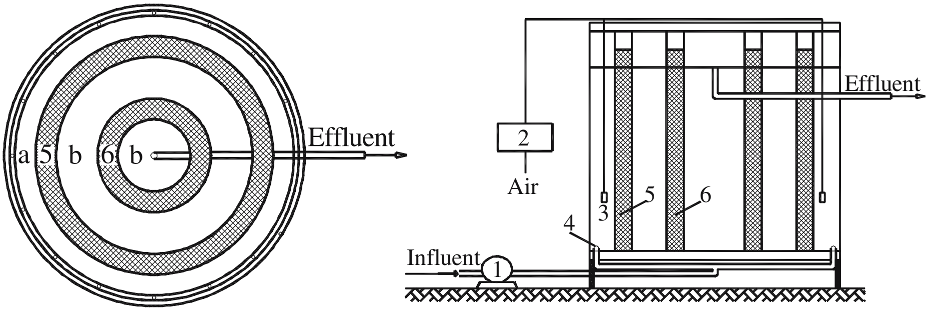

Fig. 1 Scheme of a radial-flow aerobic-anoxic immobilized biomass reactor (RAIB). (1) Top view: (a) aeration chamber (aerobic zone), (b) anoxic zone. (2) Lateral view: (1) diaphragm pump, (2) airflow measuring device, (3) porous aerators, (4) domestic sewage inflow, (5) ring containing aerobic immobilized biomass, (6) ring containing anoxic immobilized biomass

described by Zaiat et al. [7]. Polyurethane foam cubes (1-cm side) were used for biomass attachment in both reactors.

The RAIB reactor consisted of a 29.0-cm diameter, 20.0-cm high cylindrical PVC tube with a useful volume of 13.2 l, containing 10.0 l of liquid medium and 3.2 l of immobilization support. In the inner portion, the reactor was divided into five concentric sections separated by stainless steel screens with diameters of 7, 11, 19, and 23 cm. The aerobic section, with useful volume of 5.8 l, comprised the aeration chamber (external compartment) containing ten porous aerators arranged at the bottom, and a chamber filled with polyurethane foam cubes. The anoxic chamber consisted of three compartments, one of them filled with polyurethane foam cubes. The pretreated domestic sewage was fed into the bottom of the reactor's external section through a perforated circular distributor, which caused the material to flow radially from the aerobic to the anoxic section, and was collected at the top of the central section.

Batch Operation

In the first step, the RAIB reactor was operated in batch mode and a pseudo-first-order reaction in series model with nitrite as an intermediate [8] was adjusted to the experimental data of ammonium conversion into nitrite and nitrate.

In this phase, the temperature was kept at 22.7±0.2°C by recirculating water in a thermostat-controlled bath. Each experiment lasted for 6 h and the airflow varied from 2 to 15 l min^{-1}.

The kinetics of organic matter degradation in the batch reactor, as COD removal, was evaluated based on a first-order model with the residual concentration, as shown below:

$$C = C_{\mathrm{R}} + (C_{\mathrm{O}} - C_{\mathrm{R}})e^{-k_1^{\mathrm{app}}t} \tag{1}$$

In this expression, C_{R} is the residual concentration of organic matter (expressed as COD—mg l^{-1}), C_{o} is the initial concentration of organic matter (expressed as COD–mg l^{-1}), k_1^{app} is the first-order apparent kinetic constant (min^{-1}), and t is time (min).

Continuous-flow Operation

Initially, the airflow (Q_{air}) was set at 12 l min^{-1} and three different hydraulic retention times (HRT) were assayed, 7, 6 and 5 h, after which the 6-h HRT was adopted and airflows of 12, 10, and 8 l min^{-1} were applied. The average temperature was 22.7±0.2°C in these experiments.

Apparent kinetic constants for nitrite (k_1) and nitrate (k_2) formation of the first-order reaction in series model in each operating condition were assessed based on the mass balance in the radial reactor. For this mass balance, the reactor was assumed to behave as an ideal continuous stirred tank reactor (CSTR) in steady-state mode, resulting in the following expressions:

$$k_1 = \frac{\left[\mathrm{N}-\mathrm{NH}_4^+\right]_\mathrm{O} - \left[\mathrm{N}-\mathrm{NH}_4^+\right]_\mathrm{f}}{\mathrm{HRT}\left[\mathrm{N}-\mathrm{NH}_4^+\right]_\mathrm{f}} \tag{2}$$

$$k_2 = \frac{\left[\mathrm{N}-\mathrm{NO}_3^-\right]_\mathrm{f}}{\mathrm{HRT}\left[\mathrm{N}-\mathrm{N}_2^-\right]_\mathrm{f}} \tag{3}$$

$$\left[\mathrm{N}-\mathrm{NO}_2^-\right]_\mathrm{f} = \frac{k_1\left[\mathrm{N}-\mathrm{NH}_4^+\right]_\mathrm{f}}{\left(\frac{1}{\mathrm{HRT}}\right)+k_2} \tag{4}$$

After the nitrification process was established in the aerobic section of the reactor, ethanol was added to the first ring of the anoxic section of the reactor to trigger the denitrification process [9].

The analytical procedures followed the methodologies described by *Standard Methods for the Examination of Water and Wastewater* [10].

Results and Discussion

Batch Experiments

The initial filtered-sample COD values varied from 77 to 114 mg l^{-1}, with an average value of 97±12 mg l^{-1}. The residual COD oscillated between 39 and 68 mg l^{-1}, with an average value of 54±8 mg l^{-1}.

It is likely that the low content of organic matter exerted little influence on the nitrification kinetics. The values of the apparent first-order kinetic constant for organic matter degradation (k_1^{app}) varied from 1.58 to 2.78 h^{-1}, increasing with the injected airflow.

The initial ammonium concentration varied from 46 to 62 mg $\mathrm{N}-\mathrm{NH}_4^+\mathrm{l}^{-1}$, with an average value of 48±6 mg $\mathrm{N}-\mathrm{NH}_4^+\mathrm{l}^{-1}$. After a 6-h batch cycle, the ammonium concentration ranged from 2 to 17 mg $\mathrm{N}-\mathrm{NH}_4^+\mathrm{l}^{-1}$, with an average value of 9±4 mg $\mathrm{N}-\mathrm{NH}_4^+\mathrm{l}^{-1}$. It may be noted that even after a period of 6 h, a substantial portion of ammonium was not converted into nitrite or nitrate.

The pseudo-first-order reaction in series model with nitrite as an intermediate was adjusted to the experimental data and the kinetic constants presented average values of 0.25±0.06 and 6.62±0.66 h^{-1} for k_1 and k_2, respectively, indicating that the conversion of ammonium to nitrite was the limiting step of the process. Typical curves of adjustment of the model to the experimental data are presented in Fig. 2.

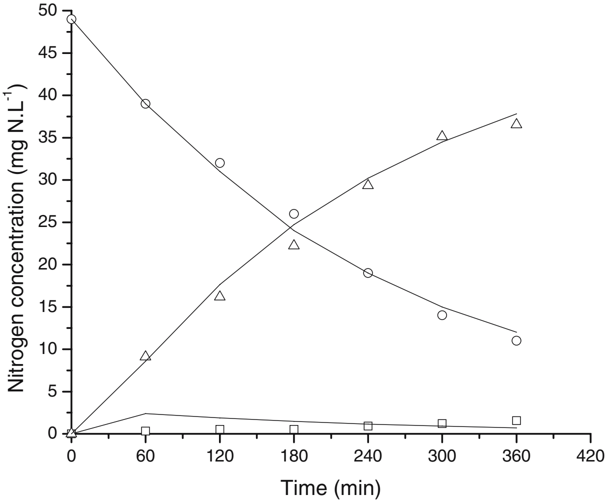

Fig. 2 Typical curves of adjustment of the first-order reaction in series model (—) to the experimental data obtained in batch experiments of *N*-ammonium (o), *N*-nitrite (□), and *N*-nitrate (Δ) concentrations

Continuous-flow Operation Experiments

Figure 3a illustrates the temporal variation of the COD of filtered samples under the reactor's various operational conditions. The COD in the effluent was usually less than 50 mg l^{-1} and the anaerobic reactor reduced oscillations observed in the organic load of domestic sewage.

Figure 3b shows temporal variations of ammonium and nitrate concentrations in the influent and effluent under the various experimental conditions. It was found that the performance of the nitrification process was considerably affected by the operating conditions. The best condition for nitrification was obtained with an HRT equal to or longer than 6 h (3.3 h in the aerobic section) and a Q_{air} of 12 l min^{-1}. When the HRT was reduced to 5 h (2.8 h in the aerobic section) with the same Q_{air}, the production of nitrate declined sharply, indicating the sensitivity of the process as a slight decrease in the HRT severely

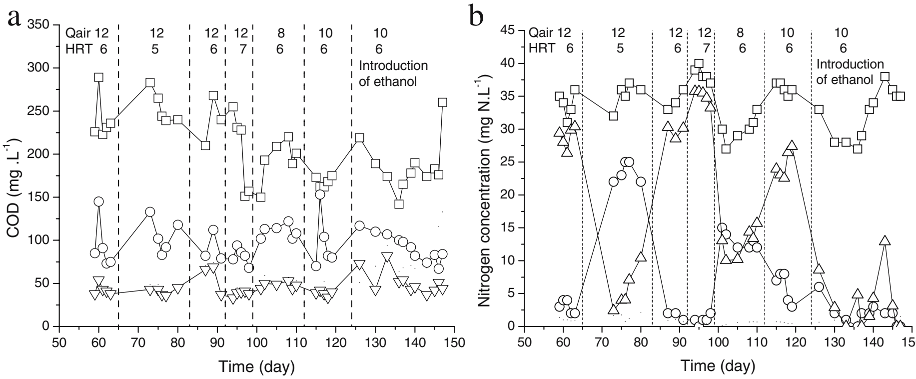

Fig. 3 (a) Values of the filtered COD in domestic sewage (□), in the influent of the RAIB reactor (○), and in the effluent (▿); (b) values of *N*-ammonium in the influent of the RAIB reactor (□), and *N*-ammonium (○) and *N*-nitrate (Δ) in the effluent of the system

impaired the nitrification process. Possibly the reduction of the hydraulic retention time might have caused an increase in the concentration of organic matter and in the content of inhibiting substances, such as sulfate. The reduction in the airflow to less than 12 l min^{-1} (8 and 10 l min^{-1}) also impaired the nitrification process. When the airflow decreased by 30% from 12 to 8 l min^{-1}, the nitrate production decreased to more than 50%, from 29 to 13 mg l^{-1}, under the same HRT. Although the dissolved oxygen reached 5 mg l^{-1} in all the experimental conditions, the oxygen transfer resistance might have been increased by the low airflow rate. It is worth mentioning that alkalinity was not a limiting factor in the nitrification process, as sodium bicarbonate was always added in excess.

After the 124th day, with the introduction of ethanol (COD/N ratio of 3.73), nitrate was removed by denitrification, providing effective nitrogen removal in the aerobic-anoxic radial reactor, besides removing the residual organic matter. In this experiment, the airflow was set at 10 l min^{-1} and an overall HRT of 6 h (3.3 h in the aerated section) was applied. Concentrations of 2 mg $\mathrm{N-NH_4^+ l^{-1}}$, 1.24 mg $\mathrm{N-NO_2^- l^{-1}}$ and 3.46 mg $\mathrm{N-NO_3^- l^{-1}}$ were obtained in the effluent of the RAIB reactor for an average influent concentration of 32±4 mg $\mathrm{N-NH_4^+ l^{-1}}$.

Table 1 lists the average values for the different forms of nitrogen in the influent and in the intermediate ring (anoxic zone) for the different operational conditions. These data refer to the effluent from the aerobic zone and indicate the capacity of nitrification. In this table was considered the hydraulic retention time effectively applied in the nitrification step, 2.8, 3.3 and 3.9 h, corresponding to the total HRT of 5, 6, and 7 h, respectively, in the reactor.

Comparing these results with those obtained in the batch experiments, it can be seen that the conversion of the *N*-ammonium in the continuous-flow system was generally more efficient, even with the shortest hydraulic retention time.

The influence of the operational conditions on the apparent kinetic constants of the first-order reaction in series model for nitrite (k_1) and nitrate (k_2) formation, Eqs. (2), (3), and (4), was adjusted to the experimental data and is presented in Fig. 4. The value of k_2 was higher than that of k_1 in all the operating conditions, indicating that nitrite formation was the rate limiting step of the nitrification process, as observed previously in the batch experiments. However, the k_2/k_1 ratio decreased sharply as the airflow increased from 8 to 12 l min^{-1}, as the values of k_1 dropped from 4.83 to 0.34 h^{-1}, whereas k_2 presented a mean

Table 1 Descriptive statistics of the nitrogen (mg N l^{-1}), considering the different operational conditions in the RAIB reactor.

Phase	Description	*N*-ammonium influent	*N*-ammonium anoxic zone*	*N*-*n*itrite anoxic zone*	*N*-nitrate anoxic zone*
Q_{air}=12 l min^{-1}	AVG±STD	35±2	23±2	1.50±0.50	5.39±3.14
HRT=2.8 h	*N*	5			
Q_{air}=12 l min^{-1}	AVG±STD	34±2	2±1	1.01±0.25	28.66±1.81
HRT=3.3 h	*N*	8			
Q_{air}=12 l min^{-1}	AVG±STD	39±1	1±1	0.51±0.07	34.69±1.32
HRT=3.9 h	*N*	5			
Q_{air}=8 l min^{-1}	AVG±STD	30±2	14±2	0.53±0.10	12.70±1.85
HRT=3.3 h	*N*	6			
Q_{air}=10 l min^{-1}	AVG±STD	35±1	6±2	0.54±0.08	25.17±2.64
HRT=3.3 h	*N*	5			

*Section “b” in Fig. 1

N = Sample numbers, AVG = average, STD = standard deviation

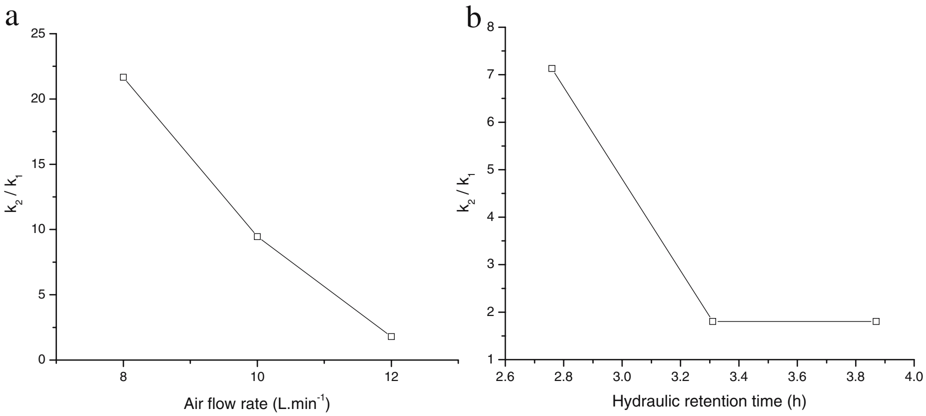

Fig. 4 (a) Variation of the relation k_2/k_1 with airflow rate; (b) variation of the relation k_2/k_1 with the hydraulic retention time applied in the aerated section

value of 9.98±3.34 h^{-1}. Hence, changes in the oxygen transfer flux affected mainly the conversion of ammonium to nitrite.

The HRT in the aerobic section of the reactor also affected the k_2/k_1 ratio. Apparently, the increase in HRT provides a better equilibrium between organisms that convert ammonium to nitrite and microorganisms that convert nitrite to nitrate, so that the k_2/k_1 ratio remained constant during the longest hydraulic retention time. In this case, both k_1 and k_2 decreased along with the decreasing HRT. Among other factors that might have contributed to the reduction of the kinetic constant values were the competition with heterotrophic organisms and the presence of inhibiting substances such as sulfate, particularly in the first minutes of the experiments. Experiments carried out by Aesoy et al. [11] demonstrated that the presence of sulfates and organic matter (less than 215 mg l^{-1} as COD) in septic sewage may reduce the nitration capacity in a biofilm system by 30 to 40% in comparison with anoxic sewage obtained by the addition of nitrate. The authors inferred that the concentration of sulfates in the proportion of 0.5 mg l^{-1} exerted a considerably negative effect on the nitrification kinetics. Cybis et al. [12] reported a reduction in the oxygen consumption rate by aerobic organisms as the concentration of sulfate in anaerobically pretreated effluent increased.

Conclusions

The results obtained during the development of this experiment allowed us to conclude that:

- The combined HAIB–RAIB system was a feasible technology to treat domestic sewage.
- Simultaneous nitrification and denitrification was achieved in a the radial-flow aerobic-anoxic immobilized biomass reactor, producing the effluent with the average concentrations of $2\ \mathrm{mg\ N - NH_4^+ l^{-1}}$, $1.24\ \mathrm{mg\ N - NO_2^- l^{-1}}$ and $3.46\ \mathrm{mg\ N - NO_3^- l^{-1}}$.
- The mass transfer resistances and the presence of inhibiting substances such as sulfates possibly interfered the *N*-ammonium conversion, as the dissolved oxygen concentration and the alkalinity were not limiting factors in the liquid medium.

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