Denitrification efficiencies of the industrial wastewater in fluidized bed bioreactors

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Abstract

A fluidized bed column was used to determine the removal efficiency of $N\text{-}NO_3^-$ in the denitrification process of wastewater. Sand with the mean particle diameter of 0.26 mm and the unexpended bed porosity of 0.43 were selected as a support media for biofilm growth. A void fraction of 0.72 was maintained during all experiments by recirculation of the effluent with a superficial velocity of 30 mh^{-1}. The pH value was controlled in the range of 7.9 ± 0.4. The other conditions of reaction varied as follows: t = 19.5 – 20.8°C; dissolved oxygen concentration (DO) = 0.15 – 0.45 mg/l. For the applied nitrate load up to 10.7 kg $N\text{-}NO_3^-$ m^{-3} d^{-1} the obtained nitrate reduction was around 98%.

Keywords: denitrification, attached biofilm growth, fluidized bed.

Introduction

The biological denitrification is one of most selective and performant methods for nitrate removal from wastewater by bacterial conversion to gaseous nitrogen [1].
The most important requirements of wastewater treatment are the high efficiencies for the denitrification bioreactors and a maximal quality of the effluent. The first aspect directly depends by the used type of bioreactor and by the methods for the acceleration of mass transfer. The second aspect depends on the reaction conditions: pH, D.O., temperature, COD/$N\text{-}NO_3$ ratio [2, 3].

Fluidized bed reactors, provide hydrodynamic conditions for the acceleration of mass transfer processes involved in biological denitrification method [4].

The aim of this study is to establish the rate of biological denitrification process using a fluidized bed bioreactor keeping the process parameters in the range mentioned by data in the literature.

Materials and methods

Experimental installation

The pilot laboratory installation used in this study, presented in Fig. 1, consists of one column with 0.06 m inner diameter and 1.50 m height. A sand fraction with particle diameter in the range of 0.1 – 0.385 mm and specific weight of 2500 kg m^{-3} was used as support. The height of the unexpanded bed put into the column was 0.5 m and its void fraction was 0.43. The experiment was conducted in a fluidized bed with the height of 1.00 m and a corresponding void fraction of 0.72. A recirculation peristaltic pump was used both to maintain the height of the fluidized bed and for the control of biofilm growth. The hydraulic load applied in the column, in order to obtain a 1.00 m height fluidized bed, was 30 mh^{-1}. Sand particles, from the fluidized bed, have been inoculated, for a period of 14 days, with biological sludge sampled from a municipal wastewater treatment plant.

Selection and growth of heterotrophic bacterial culture took place during that period, in controlled anoxic conditions. $N\text{-}NO_3^-$ and COD analyses were performed during the inoculation stage.

Nitrate concentration was daily corrected to about 10 mg $N\text{-}NO_3^-$ L^{-1}, by addition of KNO_3. An COD/ $N\text{-}NO_3^-$ ratio of 4/1 – 6/1 was maintained, adding of necessary CH_3COONa quantities. The pH was adjusted manually in the range of 7.5 – 8.5 and temperature varied in the range of 19 – 22°C.

After inoculation stage, the bioreactor was continuously fed with synthetic influents, obtained from municipal wastewater and increasing amounts of KNO_3 and CH_3COONa. Applied loads varied from 0.07 up to 10.7 kg $N\text{-}NO_3^-$ m^{-3} day^{-1}. COD / $N\text{-}NO_3^-$ ratios varied in the range of 3.5/1 – 6.3/1.

In order to allow the biomass growth, PO_4^{3-} was added using a N:P = 5:1 ratio. During the experiment, pH was controlled in the range of 7.9 ± 0.4. Solutions of sulfuric acid with 0.05 N – 0.5 N concentration were used as correction reagent.

The concentration of dissolved oxygen and water temperature varied between 0.15 – 0.45 mg O_2 L^{-1} and 19.5 – 20.8 ˚C respectively.

Analytical methods

The concentration of dissolved oxygen was measured using a Multiline F/SET3 apparatus with CellOx 325 electrode (produced by WTW – Germany). The pH was automatically adjusted using a control circuit with a GLI International electrode (USA).

NO_3^-, NO_2^- and PO_4^{3-} influent and effluent concentrations were measured using a UV-VIS 205 spectrophotometer (produced by Analytik Jena – Germany) by the following methods:

- for NO_3^- - SR ISO 7890-1 / 1998
- for NO_2^- - SR ISO 677 / 1996
- for PO_4^{3-} - SR EN 1189 / 2000

COD, both for influent effluent, was measured by volumetric method using SR ISO 6060 / 1996 method. The fluidization flows were measured using an F-460 type flowmeter for liquids (produced by Cole Parmer – USA).

fig 1

Results and discussions

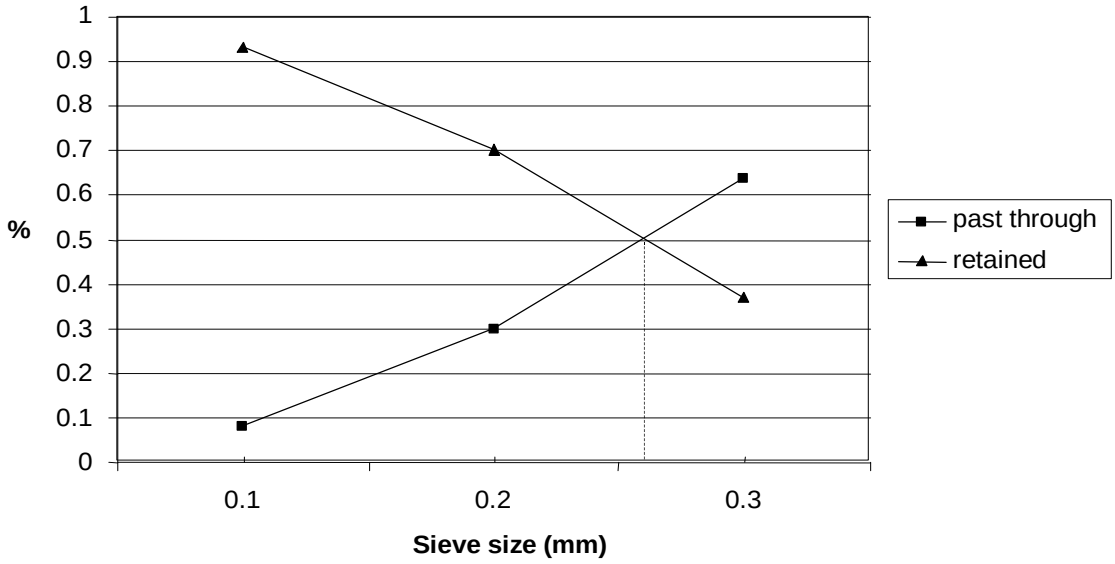
The hydrodynamics of biphasic fluidized bed

The hydrodynamics of biphasic fluidized bed is determined by physical properties of the phases (liquid specific weight and viscosity, particle specific weight, size and geometry), operating parameters (liquid flow, unexpanded bed height) and reactor size (column diameter).

The main hydrodynamic parameters considered for fluidized bed were: minimum fluidization velocity, transport velocity, bed porosity and specific surface for biomass growth ensured by the fluidized bed.

Particle average diameter was graphically determined at the intersection of "past through" and "retained" curves on sieves with ĺ0.1, ĺ0.2, ĺ0.3 mm size, as shown in Fig. 2.

Figure no 2 Mean particles diameter



A mean value for the particle diameter of 0.26 mm resulted. Shape factor for support particles, calculated [4] with the formulae $\varepsilon_0 = (0.083\ \psi)^{1/3}$ where $\varepsilon_0=0.43$ is the void fraction of unexpanded bed resulted in a value of $\psi=0.95$.

Minimum fluidization velocity, calculated [4] with Rowe equation, had a value $w_m=2.63\ mh^{-1}$. Maximum fluidization velocity / transport velocity calculated with the equation of sedimentation velocity [4] for the minimum diameter of sand particles ($d_p=0.11$ mm) and $1<Re<3000$, lead to a value of $w_t=42\ m\ h^{-1}$.

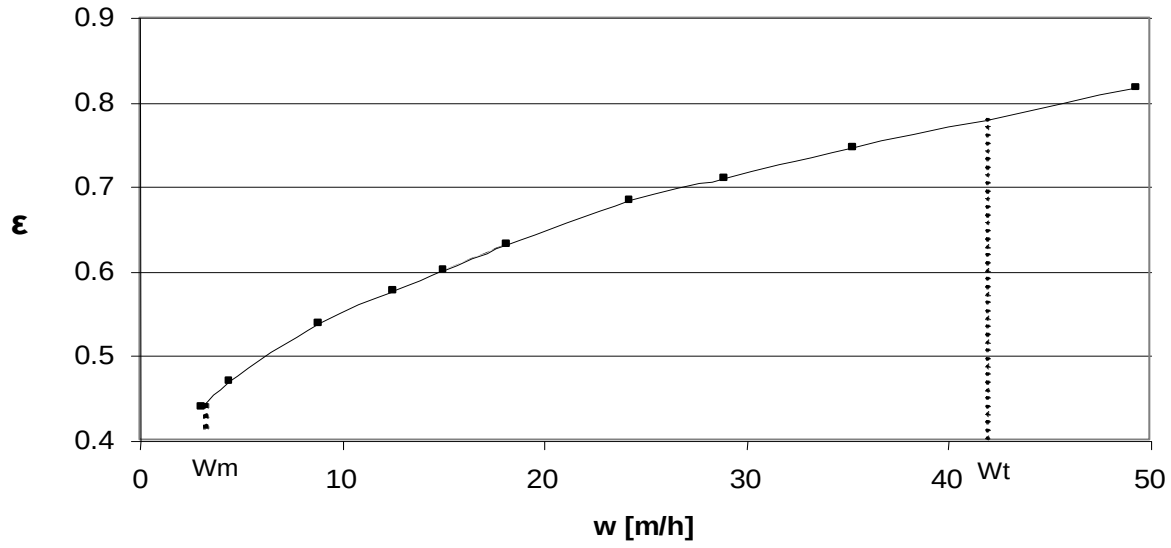
Fluidized bed porosity was experimentally determined for increasing values of applied hydraulic loads.

The results are presented in Tab. 1 and Fig. 3.

Table 1 Fluidized bed porosity

w [m h^{-1}]	1.80	3.08	4.41	8.81	12.53	14.97	18.15	24.21	28.88	32.25	49.26
H/H_0	1	1.02	1.08	1.24	1.35	1.43	1.55	1.81	1.97	2.26	3.14
ε_w	0.429	0.440	0.471	0.540	0.577	0.601	0.632	0.685	0.710	0.747	0.818

Fig. 3 Fluidized bed porosity



For a 30 m h^{-1} value of the superficial velocity in the column, the fluidized bed porosity was 0.72. Specific surface, in the fluidized bed with a void fraction of 0.72, was calculated [2] with the formulae $\sigma = 6(1-\varepsilon)\ \psi^{-1}\ đ^{-1}$, resulted a value of about 7000 $m^2\ m^{-3}$. With the dimensions of working fluidized bed, a value of about 20 m^2 available for biofilm growth is obtained.

During the denitrification experiments it was noted that in the same time with $N\text{-}NO_3^-$ load increasing, gas bubbles (N_2 and CO_2) appeared in the reaction mass become bigger and more frequent.

Therefore, the initial biphasic fluidized bed becomes a three-phase system. The gas phase resulted from denitrification reaction and the decrease of bio-particle specific weight produces an increase of fluidized bed height. This increase can be controlled by reduction of hydraulic load, in a first phase, and then by using the pump for the control of biomass growth,

which is responsible for biofilm detachment from the support particles returned from the upper part of the fluidized bed.

Microorganisms

The microscopic analyses of activated sludge suspension shows, during the first half of inoculation stage, a large number of free non-flocculated bacteria which is decreasing at the same time with the growth of small ciliates bacteria: *Colpoda sp., Colpidium sp., Glaucoma sp*.

In the second week of inoculation, fixed ciliates (*Vorticella campanula*) and suctors (*Podophrya*) were identified. At the same time the effluent becomes more transparent. At the end of inoculation stage, peritriches appeared. *Vorticella microstoma* was identified.

Biofilter support particles inoculation was considered finished when a growth of support attached biomass was observed correlated with effluent transparency, reduction of organic load and reduction of nitrate nitrogen load.

During the first days of experiment performed in continuous system, a large variety of flagellates *(Bodo, Peranema*) and rarely free small ciliates (*Colpoda, Colpidium, Uronema*) was observed. In the following days, both flagelates (*Bodo, Peranema*) and rarely free small ciliates (*Colpoda, Colpidium, Uronema*) were observed.

In the following days, both flagellates and ciliates were replaced by *Vorticella* and carnivore ciliates, (*Litonotus* and *Amphileptus).*

Nitrate removal rate

Denitrification experiments were carried in a installation fluidized bed biofilter, including a secondary settling tank and a recirculation pump, for a period of 18 days. The bioreactor was fed continuously with influent containing increasing quantities of nitrate.

Daily increasing rate for nitrate load, applied to the biofilter, was up to 50 %, as was shown in Tab. 2.

In the same time, the doses of organic carbon and phosphorus were increased adding CH_3COONa and KH_2PO_4 respectively. COD/N-NO_3^- ratio was maintained at 3.5/1 – 6.3/1 and N:P ratio at 5:1.

Table 2 Denitrification rate

Day of expts.	$N\text{-}NO_3^-$ [mg L^{-1}]		$N\text{-}NO_2^-$ [mg L^{-1}] Effluent	η [%]	Denitrification rate kg $N\text{-}NO_3^-$ m^{-3} day^{-1}	COD [mgO_2/l]		η [%]	COD / $N\text{-}NO_3^-$ ratio
	Influent	Effluent				Influent	Effluent		
1	11.2	4.4	0.11	60	0.043	57	28	51	5.09
2	14.4	3.8	0.11	73	0.057	74	30	59	5.14
3	22.1	5.4	0.15	75	0.090	104	36	65	4.71
4	26.6	7.1	0.25	72	0.093	105	5	95	3.95
5	40.2	13.0	0.58	66	0.122	142	13	91	3.53
6	51.8	3.8	0.73	91	0.193	187	19	90	3.61
7	61.7	6.7	0.71	88	0.271	327	43	87	5.30
8	87.9	3.2	1.21	95	0.352	342	24	93	3.89
9	118.0	3.0	1.52	96	0.519	452	65	86	3.83
10	139.2	3.4	1.43	97	0.686	690	116	83	4.96
11	196.0	4.9	2.89	96	0.830	892	211	76	4.55
12	252.7	3.9	3.20	97	1.406	1060	216	80	4.20
13	359.0	7.9	2.92	97	1.664	1508	267	82	4.20
14	420.0	13.5	3.11	96	2.673	2020	278	86	4.81
15	653.0	9.8	3.78	98	3.959	3592	311	91	5.50
16	895.0	14.4	3.50	98	5.843	5640	352	94	6.30
17	1137.0	31.1	3.05	97	8.175	6140	570	91	5.40
18	1259.0	22.0	3.10	98	10.505	6290	644	90	5.00

Due to the high load of nitrates applied to the biofilter, pH was kept in the recommended range (7.5-8.3) in automatic way. With the exception of the first 7 days of experiment, the removal rate for $N\text{-}NO_3^-$ was higher than 95 %, reaching 97 - 98 % for the highest applied loads. The obtained denitrification rate was up to 10.5 kg $N\text{-}NO_3^-$ per m^3 of fluidized bed and day. In the same time, the removal rate for organic load was up to 90 – 91 %, for the highest applied organic loads.

Conclusions

The use of fluidized bed technique, in the process of denitrification of wastewater with high $N\text{-}NO_3^-$ loads, conducted to $N\text{-}NO_3^-$ removal rates of 10.5 kg $N\text{-}NO_3^-$ per m^3 of fluidized bed and day. Denitrification efficiencies reached values of 95 – 98 %. The organic load needed for denitrification process was ranging between COD / $N\text{-}NO_3^-$ = 4/1 – 6/1. pH was automatically controlled in the optimum domain of 7.5 – 8.3.

At high nitrogen loads applied to the bioreactor, the biphasic fluidized bed was changing into a three-phase fluidized bed due to gaseous reaction products.

In order to stop the bio-particles to leave the reactor, a control of biomass growth is requested, using equipment able to perform the detachment of biofilm from the bio-particles situated at the upper part of fluidized bed.

The use of fluidized bed technique in the biological treatment processes for wastewater, offers two main advantages: (1) high specific surface for biomass growth and (2) hydrodynamic conditions to intensify mass transfer processes for the reagents inside the biofilm and reaction products outside the biofilm. These advantages lead to a smaller reaction volume and investment cost.

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