Journal of Cleaner Production 127 (2016) 573–578

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Note from the field

Removal of nitrate nitrogen and chemical oxygen demand in upflow fixed bed reactor using heterotrophic microorganisms

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ARTICLE INFO

Article history:
Received 30 June 2015
Received in revised form
22 March 2016
Accepted 29 March 2016
Available online 9 April 2016

Keywords:
Wastewater treatment
Denitrification
Kinetics
Monod
Statistical study

ABSTRACT

This communication deals with the removal of contaminants namely nitrate nitrogen ($N-NO_3^-$) and chemical oxygen demand (COD) in the form of acetate using an upflow packed bed anaerobic reactor. Effects of various parameters such as nutrient concentration, time, pH and flow rate were studied. The contaminants removal efficiency increased with the increase of initial concentration while the reaction rate of contaminants decreased with the increase of initial pH beyond 7. The effluent flow rate also showed a positive correlation with the contaminant removal rate up to 24 mL/min and beyond that it showed a negative trend. The reaction followed pseudo first order rate and using the same a unified rate equation was developed for both the contaminants such as $N-NO_3^-$ and COD. The mass transfer rates for the both the contaminants were calculated. Rate equations were also developed using the sorption model, which showed good agreement with the experimental results. Using Monod equation, maximum specific degradation rate as well as half saturation constant for both the contaminants was calculated. Statistical techniques including principal component analyses and multi linear regression analyses were carried out, which substantiated the analysis of the results further.

1. Introduction

Water, soil and air are three main ingredients to support the life system (Capra and Scicolone, 2007). The three ingredients need to be maintained properly to keep the fragile ecosystem under control. Rapid growth of population with commensurate growth of industry, infrastructure and agricultural is resulting in a progressive depletion of the quality of air and water. Water pollution is one of the major environmental issues and has become a global problem (Igos et al., 2014). Wastewater is mainly associated with various nitrogenous and organic compounds. Nitrogen wastes when released in water bodies lead to eutriphication (Saeed and Sun, 2012) and undesirable environmental changes that is global warming and ground water pollution (Show et al., 2013). Organic material, on the other hand decreases the dissolved oxygen, which is an essential component to support the aquatic life (Chen et al., 2011). Striving towards a more sustainable and cleaner production technique is important both from a competitive as well as in an environmental perspective (Lundkvist et al., 2013).

In the fixed film fixed bed upflow reactor, mass transfer limitation plays an important role in the reactor performance (Subbarao, 2008). The reaction rate mainly depends on the substrate as well as mass transfer either through bulk or the thin film layer surrounding the biofilm or both. A number of research papers have been published dealing with development of mathematical modeling equations for interpreting the results for biofilm reactor. Kordakandi and Berardi (2015) used artificial neural network, evolutionary polynomial regression and modified Stover Kineannon techniques to interpret the results. Vannecke et al. (2014) in depth analyzed the activities of ammonium as well as nitrite oxidizing bacteria using inverse turbulent bed reactor. Neural network technique was used to interpret the kinetics in a moving bed sequencing batch biofilm reactor (Dulkadiroglu et al., 2015). Simultaneous nitrification and denitrfication studies were also carried out in a single three phase fluidized bed biofilm reactor (Selfi and Fazaelipoor, 2012). Nitrification and dentrification studies were also carried out in moving bed biofilm reactor-membrane

Abbreviations: COD, Chemical Oxygen Demand; DO, Dissolved Oxygen; MLRA, Multi Linear Regression Analysis; PCA, Principal Component Analysis.

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http://dx.doi.org/10.1016/j.jclepro.2016.03.156
0959-6526/

reactor and pyrosequencing technique was used to analyze the activities of the nitrifying and denitrifying bacteria (Leyva-Diaz et al., 2015). Partial nitrification coupled with denitrification studies were modeled using GPS-X software (Zeng et al., 2015). Hydrodynamic and biokinetic studies were also carried out using GPS-X software (Zeng et al., 2013). One dimensional Wanner-Gujer model was used to interpret the results for nitrification process considering only suspended biomass (Masic and Eberl, 2014). The literature is scanty on kinetics as well as on identifying the rate determining step. Keeping in view of the above, an attempt has been initiated in this manuscript to evaluate the kinetics as well as to determine the mass transfer coefficient. Attempts have also been made to interpret the results using statistical techniques such as multi linear regression analysis (MLRA) and principal component analysis (PCA).

2. Materials and method

The upflow fixed bed reactor was fabricated using a cylindrical PVC pipe. The height and diameter of the reactor was 58 cm and 7.5 cm respectively. The bioreactor was packed with 3.2 kg of gravel. The gravel size was in the range of 0.2–0.5 cm. The fine particles were sieved out in order to avoid the chocking during the operation of the reactor. The void volume of the reactor was found to be 1 L. The aspirator bottle was connected with the reactor through peristaltic pump (Miclins peristaltic pump, PP 20 EX-2C make) with proper silicon tubing. After passing through the reactor the effluent was put into the same aspirator bottle. The total system was made air tight to ensure proper anaerobic conditions. Before carrying out the actual treatment experiments fixed biomass film was created. The biomass was isolated from the soil sample using enrichment technique. The denitrifying heterotrophic bacteria were isolated from the agricultural soil by MSN (mineral, salt, nutrient) liquid media (Composition: CH_3COONa-7.85 g/L, KH_2PO_4-0.2 g/L, $(NH_4)_2SO_4$-0.5 g/L, $MgSO_4.7H_2O$-0.04 g/L, $Ca(NO_3)_2$-0.04 g/L). After growth, the same was fed to the aspirator bottle along with the nutrients. The solution was circulated through the reactor till a steady state was obtained indicating proper attachment of the biomass.

Effects of various parameters such as flow rate, pH and concentrations of $N–NO_3^-$ and COD were studied. Samples were collected at regular intervals for the analyses of COD, $N–NO_3^-$, $N–NO_2^-$, dissolved oxygen and pH. The pH was measured using the Eutech Instruments Cyber Scan pH 150 make pH meter. The concentrations of COD, $N–NO_3^-$, $N–NO_2^-$ and dissolved oxygen (DO) were analyzed using APHA method (APHA, 2005). All experiments were carried out in triplicate and average of the same was used for interpretation of results. The experimental error was within ±5%. Unless or otherwise specified all experiments were conducted under following conditions: $N–NO_3^-$ 200 mg/L, COD 500 mg/L, initial pH 7 and effluent flow rate 8 mL/min.

PCA and MLRA were carried out using the SPSS-13 statistical software package.

3. Results and discussions

3.1. Variation of NO_3^- concentration, flow rate and pH

A series of experiments were carried out by varying the $N–NO_3^-$ concentration from 50 mg/L to 200 mg/L. In all the cases, acetate was used as an electron donor to reduce NO_3^-. The flow rate and initial pH were maintained at 6 mL/min and 6.5 respectively. The results are shown in Fig. 1. The nutrients depletion rates are shown in Table 1. The pH in all the cases increased with time as

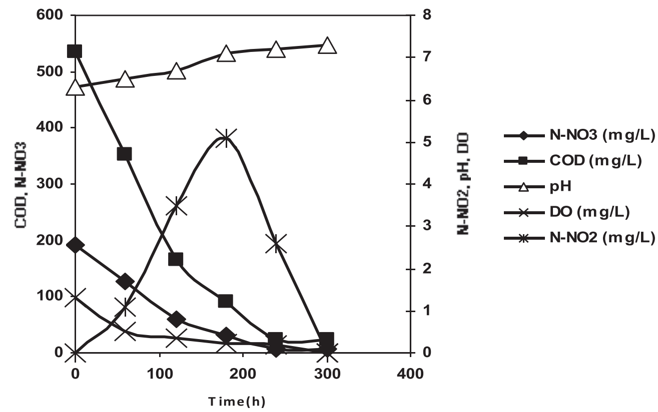

Fig. 1. Behavior of variables including $N–NO_3^-$, COD, $N–NO_2^-$, DO, pH and time during $N–NO_3^-$ variation study. (Initial conditions: $N–NO_3^-$ – 200 mg/L, Flow rate-8 mL/min, pH-6.5).

the reaction is acid consuming in nature (Park and Yoo, 2009), as shown in the reaction below.

$$0.82CH_3COOH + NO_3^- \rightarrow 0.07C_5H_7NO_2 + HCO_3^- + 0.3CO_2 + 0.9H_2O + 0.47N_2 \quad (1)$$

The DO of the solution was always less than 0.2 mg/L through out the experiments indicating good maintenance of anaerobic condition. Initially NO_2^- was non detectable but with time it increased and after around 100 min it showed a negative trend. Similar results were reported previously (Ray et al., 2014).

The effluent flow rate was varied from 6 mL/min to 40 mL/min. The $N–NO_3^-$ removal efficiency increased with an increase of flow rate till 24 mL/min and on further increase of flow rate the removal rate for $N–NO_3^-$ as well as COD decreased as shown in Fig. 2 and Table 1.

The initial pH was varied from 6.5 to 8. The contaminant reduction rate increased with the increase of pH up to 7 and on further increase it showed a decreasing trend as shown in Fig. 3. The depletion rates of contaminants are shown in Table 1.

3.2. Kinetic studies

3.2.1. Evaluation of order of reaction

The determination of the order of reaction is an important parameter to throw light on the reaction mechanism. For this purpose, two different orders of reactions such as first and second order were considered in the present case. A reaction would be considered as first or second order if linearity is obtained between a plot of ln C (C: concentration) of reactant or product versus time or 1/C versus time respectively. The results are shown in Table 1. By considering the co-efficient of determination values it can be concluded that in all the cases for all the variables the denitrification as well as COD depletion rate followed first order reaction rate. Since both of them depend on the initial concentration of the pollutant, flow rate and pH, reaction may be considered as pseudo first order (Levenspiel, 1991). As pollutant removal rate depends on three variables such as initial concentration, flow rate and pH, the rate equation can be written as

$$Rate = R = -\frac{dC}{dt} = k_1(Concentration)^{n_1}(Flowrate)^{n_2}(pH)^{n_3} \quad (2)$$

Where k_1 is the specific reaction rate constant for unified rate equation.