HIGH NITRATE WASTEWATER TREATMENT USING BIOFILTER WITH NATURAL MEDIA

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ABSTRACT

The efficiency of three laboratory scale biofilter reactors (0.748 L) packed with natural media for nitrate removal was investigated in this study. Natural materials i.e. peanut husk, sponge gourd and water hyacinth stalk were used as packing media. Ethanol was used as a carbon source in the synthetic wastewater at the concentration of 400 mg COD/L while 50, 100 and 200 mg N/L of nitrate was also added to the synthetic wastewater. This yields COD/NO_3^- ratios of 8.0, 4.0 and 2.0, respectively. The biofilters reactors were operated by semi-continuous mode with daily feeding at a constant flow rate 1.1 ml/min and hydraulic retention time (HRT) of 0.33 day. Nitrate removal efficiency of more than 80% was observed from all three media at every COD/NO_3^- ratio, except at COD/NO_3^- ratio of 2.0 in biofilter reactor packed with peanut husk. The highest nitrate removal of 98.96% was found in reactor with sponge gourd media at the COD/NO_3^- ratio of 8.0. The degradation rate was followed first order kinetic ($K= 0.1464\ d^{-1}$) and COD consumption per nitrate removed of 7.18 g COD/g NO_3^- was achieved. Results from scanning electron microscope (SEM) revealed that high porous structure of these natural media in biofilter reactor providing microorganism attachment which resulted in high COD and nitrate removal efficiency. The amount of microorganism attached on sponge gourd, water hyacinth stalks, and peanut husk was 71.3, 63.0, and 39.5 g/g dry weight, respectively. Thus, these natural media can be used in the biofilter reactor to replace the synthesized media which has higher cost.

1. INTRODUCTION

Wastewaters discharged from several industries (e.g. synthetic fibers, mineral processing, metal finishing etc.), agriculture (extensive use of chemical fertilizers) and domestic always found high concentration of nitrate. Nitrate is a common contaminant that can cause health problems in infants and anima ls, as well as the eutrophication of water bodies (Fennesy and Cronk, 1997). Physico-chemical processes such as reverse osmosis, ion exchange, and electrodialysis have been developed for nitrate removal from water but they are very expensive for pilot scale operation with a limited potential application (Kapoor, and Viraraghavan, 1977). Biological denitrification is an efficient process for nitrogen removal from wastewater in which heterotrophic bacteria in the absence of oxygen (anoxic conditions) convert nitrate and nitrite to nitrogen gas (van Rijn et al., 2006). This process requires sufficient organic carbon as an electron donor and optimum C/N ratio for complete nitrate removal.

Biofiltration is currently the most used biological treatment technology. It involves a filter bed of organic matter serving as carrier for microorganisms. (Ramírez-López, et al., 2003). According to Clark and Wnorowski (1991) almost all organic compounds can be used as biofilter carrier. In this study, three agricultural byproducts locally available in Thailand; peanut husk, sponge gourd, and water hyacinth which were used as potential carrier in biofilters. The aim of the present study is to investigate the performance of the biofilter on nitrate removal of synthetic wastewater under various COD/NO_3^- ratios.

2. EXPERIMENT

2.1 Experimental Apparatus

Three acrylic lab-scale biofilter reactors (diameter 6.9 cm; height 20 cm) with 0.748 L of total volume and

an operating volume of 0.524 L were used in this study (Fig.1). Each column was packed with natural media (size ~ 1 cm^3) i.e. peanut husk, sponge gourd, and water hyacinth. The packed height of media was 14 cm. Sampling ports were provided at the middle and the

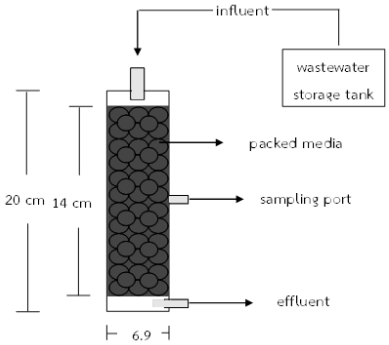

Fig. 1 Schematic diagram of a biofilter for nitrate removal used in this study.

bottom of the biofilter. All media were oven dried at 70°C and soaked in distilled water for 24 h before filled in the reactors. The synthetic wastewater was prepared according to Godini et al. (2010) as following (in mg/L): C_2H_5OH 400; $FeSO_4.7H_2O$ 0.85; $NaMO_4$ 0.25; $MnSO_4.7H_2O$ 0.157; $NaHCO_3$ 33; Na_2SO_3 20; $CoCl_2$ 0.55; K_2HPO_4 0.5; KH_2PO_4 0.5. The KNO_3 concentration of 50, 100, and 200 mg NO_3^--N/L; was added in the wastewater to provide COD/NO_3^--N ratios of 8, 4, and 2, respectively. The pH was adjusted around 7.0.

2.2 System operation

Microorganism taken from the recycle sludge of a municipal wastewater treatment plant was used as seed for denitrification in the biofilter. The seed sludge was acclimatized by stepwise increases the wastewater concentration until it reached the COD 400 mg/L and NO_3^- 50 mg/L. Then, the acclimated sludge (10,000 mg/L) was introduced into each biofilter, filled up with synthetic wastewater and allowed to remain static (no flow) for 2 weeks. After that, the synthetic wastewater was daily fed via peristaltic pumps from the top of the biofilter in semi-continuous mode (8 h/d) at a flow rate of 1.10 mL/min. The COD/NO_3^- ratio initially employed was 8, using ethanol as carbon source. Each test was performed until steady state conditions were reached, showed by constant values of both effluent COD and nitrate concentration. Samples were collected from influent, effluent and sampling ports every 24 h to determine the biofilter performance.

3. ANALYSIS

COD, Alkalinity, NO_2^-, NO_3^-, and suspended solids (SS) were analyzed according to APHA Standard Methods (2005). Oxidation–reduction potential (ORP) and pH were monitored using an ORP meter (WTW pH 315i, Electrode SenTix ORP) and a pH meter (WTW Series, pH720 InoLab), respectively. Dissolved oxygen was measured with a DO meter (WTW OX 315i, WTW Durox). Statistical analyses were carried out with data from each loading rate at steady-state conditions.

At the end of each experiment, the biomass was washed off from the media with distilled water and SS was determined for calculating g biomass / g dry media. Peanut husk, sponge gourd, and water hyacinth before and after treatment of nitrate synthetic wastewater were prepared according to Pathan, et al. (2008) and sent to analyze at Scientific and technological research equipment center, Chulalongkorn University.

4. RESULTS

Fig. 2 and Table 1 show the results obtained in the denitrification of synthetic nitrate wastewater for the different COD/NO_3^- ratios studied. Each time point is the average of triplicate measurements and three profiles were made on separate days for each experimental COD/N ratio.

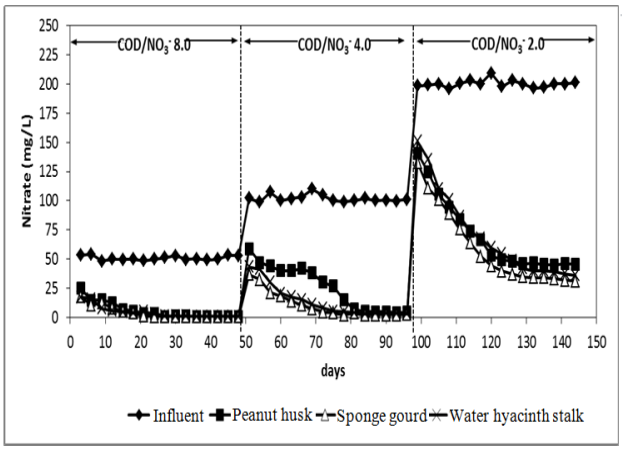

Fig. 2 Time course of influent and effluent nitrate concentration in each biofilter packed with different natural media.

Table 1 Nitrate removal efficiency, COD removal efficiency, and K_1 of biofilter packed with different natural media.

Natural media	Influent NO_3^-N (mg/L)	Nitrate Removal (mgNO_3^--N / g media)	K_1 (d^{-1})	COD removal (mg COD / g media)
Peanut husk	50	1.44	0.1262	9.22
	100	2.73	0.0643	10.27
	200	4.36	0.0431	9.13
Sponge gourd	50	9.13	0.1464	65.52
	100	17.81	0.1013	63.34
	200	30.00	0.0511	63.34
Water hyacinth	50	2.93	0.0820	18.88
	100	5.70	0.0900	21.22
	200	9.38	0.0444	19.99

For the lower ratios employed, deceleration of the denitrification rate was observed as the nitrate concentration and COD decreased below a certain level. It can be seen that all COD/NO_3^- studied ratios gave high nitrate removal efficiency higher than 80%, except COD/ NO_3^- of 2.0 with peanut husk as a packing media. The effluent pH values throughout the study were in the range of 6.89-7.44 which is suitable for denitrification process. It was recommended that the pH values should be in the range of 6.0-9.0 to avoid the microbial toxicity which will affect nitrate conversion to nitrogen gas (Farjado et al., 2012) as follow;

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$$

Observation of NO_2^- concentration in the reactors, it was found that the concentration was very low. Nitrite normally is not stable. Effluent suspended solids were in the range of 10-14 mg/L for bioreactor with sponge gourd, and they were 14-21 mg/L for bioreactor with peanut husk and water hyacinth stalk. Dissolved oxygen was in the range of 0.4-0.7 mg/L for all biofilters and the oxidation-reduction potential was in the range of -160 to -115 mV. Thus, the environmental conditions in the reactors were benefit for anoxic condition or denitrification process.

To study the biodegradation capability, the biodegradation kinetics was studied within 0-36 days of biological degradation times. It was found that microbial degradation of carbon and nitrogen in the reactor was followed first order kinetic. The first order constants (K_1) were determined from the plot of lnC and time, where C was the concentration of NO_3^--N at time t. The highest nitrate removal of 98.96% was found in reactor with sponge gourd media at the COD/NO_3^- ratio of 8.0 with the degradation rate of 0.1464 d^{-1} and COD consumption per nitrate removed of 7.18 g COD/g NO_3^-. The increment of nitrate concentration also reported the increment of nitrate removed per gram of media but the decrement of K_1.

Fig. 3 shows the surfaces of packing media before and after used in the reactor at 148 days. All natural media have roughness surfaces which are suitable for microbial attachment. High porous media also found for sponge gourd and water hyacinth stalk. Sponge gourd showed the highest removal efficiency comparing to peanut husk and water hyacinth stalk because it had high porosity. High porous media could provide space for microorganism attachment, consequently to enhance biodegradation and toleration of high nitrate concentration. Determination of the amount of microbial attachment onto the packing media also reported the higher amount of microorganisms with the higher porosity of media. The dry weight bacteria of sponge gourd was 71.3 mg/g dry weight, followed by water hyacinth stalk of 63.0 mg/g dry weight and peanut husk of 39.5 mg/g dry weight, respectively.

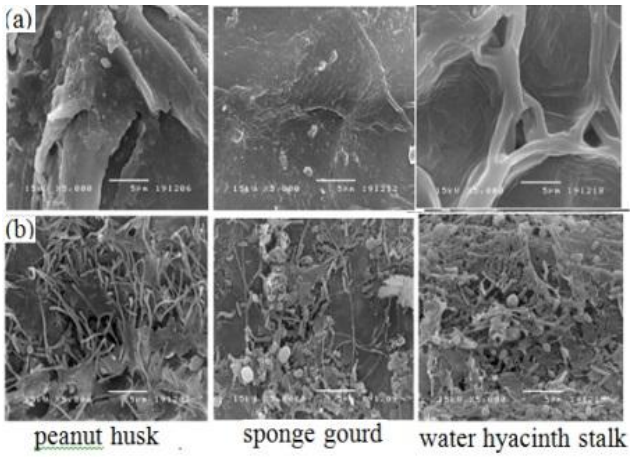

Fig. 3 Scanning electron micrograph (5000X) of the natural biofilter media surfaces before used (a) and after 148 days of wastewater treatment (b).

CONCLUSION

Natural media from agriculture by product can be used as a packing media in biofilter for high nitrate removal. All media could remove nitrate higher than 95% when COD/NO_3^- ratios of 4 and 8 were applied. Sponge gourd is a suitable media compare to peanut husk and water hyacinth stalk.

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