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Hydrogenotrophic denitrification of synthetic aquaculture wastewater using membrane bioreactor

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Abstract

A hydrogenotrophic denitrification system was evaluated in removing nitrate from synthetic aquaculture wastewater for recirculation purposes. Two membrane bioreactor (MBR) systems, namely, aeration–denitrification system (ADS) and denitrification–aeration system (DAS) were studied with 50 mg/L of influent concentrations for both organic matter and nitrate nitrogen. The DAS achieved better removal efficiency of 91.4% total nitrogen (T-N) and denitrification rate of 363.7 mg/L.day at a HRT of 3 h compared to ADS. Further, there was no nitrite accumulation in the DAS effluent. The nitrite accumulation in ADS effluent was lesser when CO_2 was used as buffer rather than K_2HPO_4 and KH_2PO_4. Estimation of kinetic parameters of hydrogenotrophic bacteria indicated lesser sludge production compared to heterotrophic denitrification. In the DAS, membrane fouling was nonexistent in the aeration reactor that was used to produce the recirculating effluent. On the contrary, membrane fouling was observed in the denitrification reactor that supplied hydrogen to the mixed liquor. Thus, this study demonstrated DAS capability in maintaining the acceptable water quality appropriate for aquaculture, in which a closed recirculating system is typically used.

Keywords: Aquaculture wastewater; Effluent re-circulation; Hollow fiber membrane; Hydrogenotrophic denitrification; Membrane bioreactor

1. Introduction

Aquaculture has been developed steadily over the last decade in response to the increasing world market demand for seafood. However, it also discharges an enormous amount of wastewater into the environment which contains high concentration of nitrites, nitrates and phosphorus, which eventually leads to eutrophication on receiving waters. Eutrophication affects benthic fauna, macroalgal growth and diversity, epiphyte communities, phytoplankton, zooplankton and bacterial communities. Concerned agencies have started issuing load-based licenses to aquaculture farmers to minimize discharge of nitrogen and phosphorous into the environment [1]. Consequently, aquaculture industries look for appropriate and better methods in treating wastewater prior to recirculation or discharge into the receiving waters.

Table 1 shows the production capacities and effluent qualities of several fresh and saline water aquaculture systems [2–5]. Ammonia produced by the animals undergoes nitrification in a recirculating system. Nitrate ion is the end product of the nitrification process and it tends to accumulate in a closed recirculating system. For high-density fish aquaculture, nitrate value can reach up to 500 mg/L as NO_3^--N in both brackish and seawater recirculating systems [6]. Similarly, a recirculating seawater system used in rearing tiger shrimp brood stock showed an accumulation of NO_3^--N up to 50 mg/L in a 40-day trial period [7]. Substituting a fraction of the water with low nitrate value water (i.e. fresh water) can reduce nitrate concentration in the system. However, this is not a suitable approach because of high cost involved in large water exchange especially in areas where water supply is limited, the environmental assimilative capacity is low and have strict legislative restrictions on effluent discharge [8,9]. Hence, biological denitrification is an alternative means to remove the nitrate. In this process, nitrate is reduced to gaseous nitrogen prior to release in the atmosphere [10–12]. In biological denitrification, organic carbon serves as electron donor for heterotrophic bacteria whereas inorganic compounds are consumed by autotrophic bacteria. Most of the water in aquaculture has low biochemical oxygen demand (BOD); hence, an external electron donor is required [13]. Traditionally, organic

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Table 1
General characteristic of production and effluent quality of aquaculture farms

	Barramundi fish [2]	Shrimp culture [3]	Shrimp culture [3]	Taiwan shrimp village [4]	Harlington shrimp farm [4]	Catfish [5]
Stocking density	65 kg/m^3	30 no/m^2	70 no/m^2	50 PL m^{-2}	15 PL m^{-2}	40 kg/m^3
Water exchange (% day)	14.3	–	–	10	7	20
Salinity (ppt)	0	–	–	12	40	–
Yield (kg ha^{-1})	–	–	–	4630	1777	–
System type	Recirculating	Open loop	Open loop	Open loop	Open loop	Recirculating
Treatment	Trickling filter	–	–	–	–	Trickling filter
Effluent quality						
pH	7.0–7.6	–	–	7.83	8.51	4.7
DO (mg/L)	–	–	–	5.31	5.38	2.5
NH_3-N (mg/L)	0.40–3.42	0.98	6.50	1.36	0.04	1.5
NO_2^--N (mg/L)	0.13–1.53	0.02	0.08	0.41	0.01	20–220
NO_3^--N (mg/L)	1.39–13.78	0.07	0.15	0.67	0.65	–
Total P (mg/L)	–	0.18	0.32	0.51	0.15	–
Total reactive P (mg/L)	0.26–3.52	–	–	0.24	0.01	–
Carbonaceous BOD (mg/L)	–	10.0	28.8	2.95	9.16	–
Total suspended solids (mg/L)	–	–	–	93.0	95.1	

electron donors such as methanol are used, where anaerobic bacteria utilize nitrate as electron acceptor under anoxic conditions. This process must be controlled carefully to avoid overdosing of organic electron which can lead to water quality degradation. Biodegradable polymers are used as organic electron donors, in which they act both as a biofilm carrier and carbon source [14]. The organic matter found in the recirculating fish culture system was also used as electron donor in the denitrification reactor. In addition, elemental sulfur has been utilized for autotrophic denitrification regardless of its drawbacks such as alkalinity consumption and sulphate production [15].

Hydrogen gas is a safe alternative to organic electron donors and element sulphur. It is non-toxic and unwanted by-products are avoided [16]. Furthermore, it is cheap and generates 50% less microbial biomass than traditional electron donors such as methanol [17]. By using hydrogen gas as the electron donor, hydrogenotrophic denitrification can occur in the absence of oxygen and nitrate is reduced to nitrogen. Several bacteria such as *P. denitrificans*, three species of *Hydrogenophaga* and *Alcaligenes eutrophus* are capable of hydrogenotrophic denitrification. *P. denitrificans* can grow autotrophically as hydrogen oxidizing denitrifier. On the other hand, *Alcaligenes eutrophus* showed slow growth [18]. Low solubility of hydrogen gas inhibits hydrogenotrophic denitrification that leads to accumulation and explosion in closed spaces [19]. Several studies have reported using gas permeable membrane in dispersing hydrogen gas into the reactor with high efficiency [20,21]. By using a bubble-free permeable membrane, delivery of hydrogen gas was successful without creating an explosive environment [17,19,22].

In 1986, a full scale plant in Mönchengladbach, Germany started using DENITROPUR to treat groundwater with 80 mg/L of nitrate [23]. The treatment capacity of the plant was 100 m^3/h. It consisted of hydrogen saturator which combined the nitrate laden water with hydrogen and allowed the mixture to pass through a number of packed-bed reactors for hydrogenotrophic denitrification. The plant eliminated 90 kg nitrate/day. Further, Grommen et al. [24] studied the removal of nitrate in aquaria using an electrochemically generated hydrogen gas as electron donor for biological denitrification. In that study, electrochemical cell was used to generate hydrogen gas. During a 7-day aquarium test, nitrate removal rate increased up to 18.5 mg N/L of reactor volume per day at an influent NO_3^--N concentration of 20 mg/L.

From the start, biological treatment system is necessary to treat the effluent from a recirculating aquaculture system to avoid treating higher concentrations of nitrate as it slowly build up inside the culture tanks. The treatment system reduces the rate of nitrate build-up which can lessen the amount of water required for freshwater exchange. Hence, this paper aims to investigate the performance of hydrogenotrophic denitrification using hollow fiber membrane in treating aquaculture wastewater needed for recirculation. Experiments were conducted in order to:

- investigate the potential of autotrophic, hydrogen oxidizing bacteria for denitrifying aquaculture wastewater;
- identify various design parameters and operational requirements which play a significant role in operation and performance of hydrogenotrophic denitrification in hollow fiber membrane bioreactor; and
- optimize the operating conditions which could project its application on a large scale.

2. Materials and methods

2.1. Sludge acclimatization

The hydrogenotrophic denitrification microorganism was acclimatized using activated sludge taken from a treated domestic wastewater. The activated sludge was added to a 4-L flask containing nitrate, bicarbonate, tap water and buffer, to which hydrogen gas was supplied through a 2 m silicone tube. The hydrogen gas pressure was maintained at approximately 1 bar in the silicone tube. Concentration of nitrate nitrogen was gradually increased from 6 to