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Demonstration of a novel ethanol-packed membrane biofilm reactor for denitrification at the Tokyo Sea Life Park

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ABSTRACT

A novel ethanol-packed membrane biofilm reactor (MBfR) was used to develop a simple denitrification technology applicable to aerobic tanks in recirculating aquarium or aquaculture systems. First, the basic properties of the module of the MBfR were assessed via a series of batch tests. The results of the batch tests indicated that (1) the module could obtain denitrifying capability after a 2-week submergence in aerated seawater without having direct inoculation onto the module, (2) the denitrification rate was proportional to the ethanol supply rate, and (3) the type of supporting medium had little effect on denitrifying capability. On the basis of these results, a module for the full-scale demonstration was made of 30 cm × 30 cm square of non-woven fabric coated with a 0.07-mm-thick PE film on the one side. An openable tap was incorporated to supply additional ethanol, and 100 mL of ethanol was packed into each module before use. Then, the appropriate number of modules was used for a full-scale demonstration in two types of recirculating aquarium systems for up to 400 days. In an aquarium rearing small fishes, the three MBfR modules submerged in the aerobic tank prevented nitrate accumulation for 77 days. However, the surface denitrification rate (0.68 gN m^{-2} d^{-1}) was smaller than the expected value, probably because the nitrate concentration was very low. In another aquarium mainly rearing lobsters, the 17 submerged modules prevented nitrate accumulation at the rate of 1.0–1.1 gN m^{-2} d^{-1} for approximately 6 months without ethanol refilling. After that, it was also observed that the dilution of ethanol decreased the denitrification rate; however, ethanol refilling resulted in immediate recovery of denitrification capability. Taken together, our results indicate that the proposed ethanol-packed MBfR could be employed as a denitrification technology for recirculating aquariums or aquacultures, and offers the practical advantage of eliminating the need for a pump for ethanol supply and an additional loop or tank exclusively for denitrification.

1. Introduction

For rearing marine creatures in recirculating aquaculture systems, it is important to control nitrogen derived from feeding and excretion. Since the ammonium and nitrite forms of nitrogen are considerably more toxic than nitrate (Camargo et al., 2005), aerobic biofiltration, which can biologically oxidize ammonium and nitrite into nitrate (nitrification), is considered to be an essential component of recirculating systems (de Graaf, 1964; Spotte, 1970; van Rijn, 1996; van Rijn et al., 2006). Although the toxicity of nitrate is lower than that of ammonium and nitrite, the accumulation of nitrate to the level of several hundred mgN L^{-1} can be fatal, particularly for juvenile fish and some invertebrates (Camargo et al., 2005), and little is known about its chronic toxicity. Therefore, for public aquariums in which a variety of marine creatures are reared, it is generally recommended that the nitrate concentration be kept below 20 mgN L^{-1} (Spotte, 1970). If natural seawater can be easily obtained, regular water change is the simplest and most common way to control nitrate concentration. However, there are often difficulties related to the direct use of the natural seawater, even for aquariums located in costal areas, including chemical pollutants, low salinity due to river water, and temperatures that are different from rearing conditions. It should also be noted that the discharge of used seawater containing high nitrate concentrations may be regulated.

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Studies on nitrate removal in recirculating aquaculture systems have been conducted since the 1970s (van Rijn et al., 2006). Generally, it is difficult to apply physical or (electro)chemical treatments alone to seawater, because their reaction specificities for nitrate are too low to practically remove nitrate under high salinity conditions. On the other hand, some microorganisms can selectively utilize nitrate as their electron acceptor and reduce it to nitrogen gas (denitrification) after dissolved oxygen (DO), the preferred electron acceptor, has been depleted. The use of an external electron donor for denitrification, similar to the treatment process for municipal and industrial wastewater, has been the focus of a number of studies into recirculating aquaculture systems since the pioneer works by Meske (1976) for freshwater aquaculture and Balderston and Sieburth (1976) for seawater aquaculture. It has been demonstrated that in seawater aquaculture systems, even high-density rearing can be achieved in recirculating aquaculture systems equipped with denitrification units (e.g., 43.4 kg m^{-3} of Japanese flounder reported by Honda et al., 1993). In addition, large-scale denitrification units have been successfully employed in seawater aquariums, including the treatment of 3 million L at the Montreal Biodome, QC, Canada (Labbé et al., 2003; Labelle et al., 2005; Dupla et al., 2006; Laurin et al., 2006), 2.9 million L at the New Jersey State Aquarium, NJ, USA (Grguric et al., 2000a), and 23 million L at the Living Seas at EPCOT Center, FL, USA (Grguric et al., 2000b). The denitrification units in these cases consisted of commonly used equipment such as a deoxygenation tank followed by a denitrification tank and a methanol supply. Another common feature is the requirement of an additional recirculating loop independent of that for nitrification, because the optimal recirculating conditions are quite different between nitrification and denitrification (van Rijn et al., 2006). Thus, it may be difficult for existing aquariums and aquacultures to implement the traditional denitrification systems because of limited space or an unsuitable configuration.

In order to address this problem, researchers have recently developed novel denitrification technologies that can dramatically reduce the size of equipment or even eliminate the need for some of it. For example, Matos et al. (2009) proposed a compact denitrification system that effectively separated denitrifying conditions from nitrifying conditions by an ion-exchange membrane. Grommen et al. (2006) have suggested that the electrochemical generation of hydrogen gas can be compact equipment to supply the electron donor for denitrification. According to Mook et al. (2012), this concept can be enhanced through the use of a denitrifying biofilm grown on the surface of the electrodes. Another example of an emerging hydrogen-based technology is the membrane biofilm reactor (MBfR). In general, denitrifying MBfRs utilize a biofilm attached to the surface of a hydrogen-permeable hollow fiber membrane (Martin and Nerenberg, 2012). Since DO cannot reach the inner layer of the biofilm, an MBfR can effectively reduce nitrate, as well as other oxyanions such as perchlorate, arsenate, chromate, and selenate (Chung et al., 2007), even under high DO conditions. Moreover, our previous studies suggested that another types of MBfR, comprising an ethanol-packed polyethylene bag and a supporting media for biofilm, also has potential as a technology for denitrification under high DO conditions (Uemoto et al., 2009; Shoji et al., 2010; Uemoto and Morita, 2010). The ethanol-packed MBfR is characterized by its electron donor supply. Since ethanol (lower alcohols) can be gradually supplied from the polyethylene film by permeation like gaseous or liquid hydrocarbons (Aminabhavi et al., 1989; Klopffer and Flaconnèche, 2001), the novel MBfR can be made from a non-porous low-density polyethylene film, which is very inexpensive compared to the hydrogen-permeable hollow fiber membrane. Although these novel denitrification technologies seem promising for recirculating aquaculture systems (Uemoto et al., 2014), very few attempts have been made at full-scale and long-term demonstrations in functioning aquariums and aquacultures.

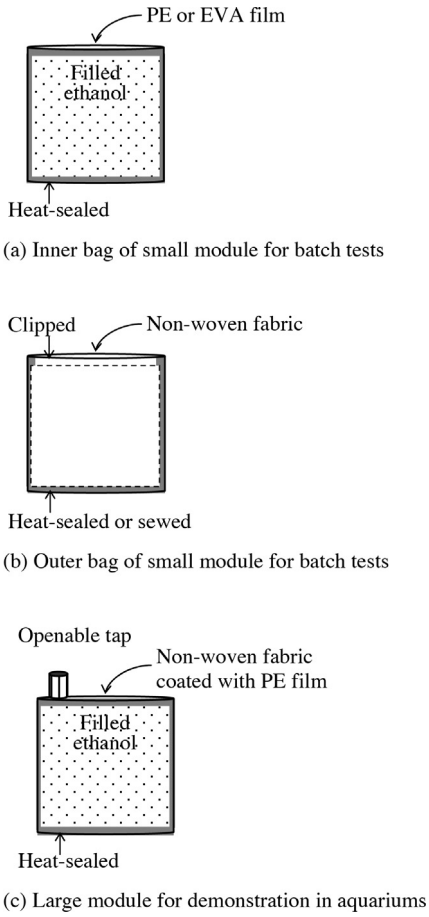

Fig. 1. Ethanol-packed MBfR modules for batch tests (a and b) and full-scale demonstrations (c).

At the Tokyo Sea Life Park, one of the largest public aquariums in Japan, there are more than 30 rearing tanks which have independent recirculating systems with aerobic biofiltration and additional equipment such as protein skimmers (foam separation) and ozonators where necessary. To prevent nitrate accumulation, regular water changes (1–5% d^{-1}) have been performed. Although the aquarium faces Tokyo Bay, seawater is obtained by truck and ship transportation because the salinity of the seawater in the immediate vicinity of Tokyo Bay is too low to rear marine creatures, as a consequence of dilution by two large rivers. Thus, compared to the present water change process, it seems quite probable that the implementation of denitrification would be a cost-effective alternative. However, because of space limitations, no traditional denitrification systems requiring additional denitrification tanks and recirculating loops have been installed. This situation has prompted the use of an ethanol-packed MBfR submerged in an existing biofiltration tank at the aquarium. The purposes of this study were to design an ethanol-packed MBfR for existing aquariums and to examine its performance during demonstrations over more than a year.

2. Material and methods

2.1. Preparation of ethanol-packed MBfR modules

The MBfR consisted of double-layer square-bag modules; the inner bag (Fig. 1a) was for ethanol supply and the outer bag (Fig. 1b) served as a supporting medium for the denitrifying biofilm (Uemoto et al., 2009). In the present study, lab-scale modules (3 cm × 5 cm)