Journal of Membrane Science 389 (2012) 67–75

Contents lists available at SciVerse ScienceDirect

Journal of Membrane Science

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

Application of a GAC-coated hollow fiber module to couple enzymatic degradation of dye on membrane to whole cell biodegradation within a membrane bioreactor

Faisal Ibney Hai[a,b,*], Kazuo Yamamoto[c], Fumiyuki Nakajima[c], Kensuke Fukushi[d]

[a] Department of Urban Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
[b] Strategic Water Infrastructure Laboratory, School of Civil, Mining and Environmental Engineering, The University of Wollongong, NSW 2522, Australia
[c] Environmental Science Center, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan
[d] Integrated Research System for Sustainability Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8654, Japan

ARTICLE INFO

Article history:
Received 1 March 2011
Received in revised form
16 September 2011
Accepted 12 October 2011
Available online 18 October 2011

Keywords:
Dye wastewater
Enzymatic membrane reactor
Granular activated carbon (GAC)
Hollow fiber module
Membrane bioreactor (MBR)
White-rot fungi

ABSTRACT

Additional granular activated carbon (GAC) layers on the membrane module within a whole cell fungal membrane bioreactor (MBR) set up to treat dye wastewater was effective in minimizing enzyme washout and in improvement of decoloration (degradation of the dye). Supporting batch tests and continuous monitoring of the quality of bioreactor-supernatant and membrane-permeate revealed that biodegradation was effected by both the suspended culture and the dynamically immobilized enzyme on the GAC-coated membrane. In a control MBR, the efficiency of dye removal was variable; in contrast, 85% to near complete removal of dye was achieved using the MBRs equipped with a membrane with additional GAC layers. A combined critical effect on dye removal efficiency of hydraulic retention time (HRT), instantaneous membrane flux and the amount of GAC coating was observed. Plausible explanations are presented for novel observations such as the achievement of better removal efficiency in the case of the longer HRT, even though the dye loading under different HRTs was kept the same by varying its concentration in the synthetic wastewater. The effect of membrane fouling on removal efficiency is highlighted and approaches to achieve stable long-term performance are discussed.

© 2011 Elsevier B.V. All rights reserved.

1. Introduction

The application of submerged membrane bioreactor (MBR) processes in municipal and industrial wastewater treatment has grown substantially in recent years owing to several advantages including excellent effluent quality, low sludge production, small foot print, robustness and flexibility for future expansion [1,2]. MBR systems are particularly attractive for treatment of recalcitrant wastewater where long sludge retention times (SRT) that facilitate physical retention and subsequent hydrolysis, are critical to achieving biological degradation of pollutants. Researchers have put forward novel modifications to conventional designs of MBR in order to enhance removal performance. Such modified designs include integrated anoxic/aerobic MBR, biofilm MBR, thermophilic MBR, bio-augmented MBR, and so on [3]. The membrane is an integral part of MBR, performing the role of solid–liquid separation, that is, they retain suspended solids (SS) as well as the soluble materials adsorbed on the SS, and thereby enhances removal compared with conventional activated sludge processes (CAS). To date, most MBR research has addressed membrane fouling mitigation (to enhance hydraulic performance of the membrane) and treatment performance improvement of the bioreactor separately. Membrane fouling mitigation strategies include modification of membrane surface and module design, alteration of mixed liquor characteristics and tuning operating conditions [4,5]. Notably, modification of surface properties of membranes to be used in MBR has been attempted mainly from the point of view of fouling mitigation [5,6].

In contrast to the membrane in a whole cell MBR, that in an enzymatic membrane reactor (EMR) is modified to act as supports for enzyme immobilization. Thus, the membrane takes part in catalytic degradation of pollutants with simultaneous downstream separation of the transformation products [7,8]. Investigations regarding application of enzymatic membrane reactors for wastewater treatment have been mainly carried out on low strength wastewater with limited total organic carbon (TOC) and total suspended solid (TSS) loadings [9]. Notably, the stabilities and catalytic properties of enzymes immobilized on membranes are dramatically affected by high strength wastewaters [10]. Gradual loss of enzymatic activity

* Corresponding author at: Strategic Water Infrastructure Laboratory, School of Civil, Mining and Environmental Engineering, The University of Wollongong, NSW 2522, Australia. Tel.: +61 2 4221 3054.
E-mail address: faisal@uow.edu.au (F.I. Hai).

0376-7388/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.memsci.2011.10.016

due to various physical, chemical and biological inhibitors under wastewater conditions is inevitable.

We envisaged that modification of membrane modules to dynamically immobilize pollutant degrading enzymes on membranes within a whole cell MBR may bring about the added advantages of continuous enzyme production (by microbes) and prevention of washout of enzyme. The resultant continuous application of enzyme, minimizing washout may mean undisrupted operation even under conditions where denaturation of enzyme would occur over time. A few studies have reported that addition of an adsorbent column downstream from the enzymatic membrane reactor resulted in improved removal performance [11]. In line with the above discussion, utilization of an adsorbent (e.g., GAC) as an enzyme immobilization support on membranes in whole cell MBR may be further beneficial for enhanced removal of contaminants from wastewater.

The proposed concept can be tested using a fungal bioreactor system which is known to suffer from unstable biological performance due to the limitations, namely bacterial contamination destabilizing fungal activity [12], and loss of the extracellular enzymes and mediators with discharged water [13]. It is worth noting that fungal reactors have been studied to find an alternative to treat various hazardous chemicals including dyes which are not degraded in bacteria-dominated conventional activated sludge. In contrast to the numerous reports on the excellent degradation capacity of pure fungus cultures or the relevant extracellular enzyme in small-scale batch-tests, there exist only a few studies which report dye degradation performance in continuous reactors [14]. Among the few studies concerning continuous fungal reactors, only a handful has explored dye degradation under non-sterile environments [14–18]. Only a few studies [13,14] so far have specifically investigated the potential of prevention of enzyme washout from continuous fungal reactors by applying simultaneous activated carbon adsorption within bioreactor. It is clear that systematic studies on enhancement of fungal degradation of dyes in continuous reactors under non-sterile environments are imperative. In this context, the proposed GAC-coated membrane may be useful.

In order to address these research gaps, the current study explored the efficacy of a GAC-coated membrane within a fungal MBR in improving performance of hazardous dye removal in wastewater treatment. Preliminary investigations showed that additional GAC layers on the membrane module enhanced decoloration and substantially decreased enzyme washout. Long-term investigations were carried out to ascertain the factors governing the performance of the membrane module. The effect of membrane fouling on removal efficiency was also highlighted. This study: (i) demonstrates the application of enzyme-immobilization on membranes within a whole cell membrane bioreactor for the first time, and (ii) suggests a potential solution to unstable removal performance of fungal reactors used for treating recalcitrant wastewater.

2. Experimental

2.1. Microorganism, chemicals and synthetic wastewater

The white-rot fungus *Coriolus versicolor*, NBRC 9791 obtained from the NITE Biological Resource Center (NBRC), Japan was used for this study. A nutrient-sufficient synthetic wastewater was prepared by adding dye (Acid Orange II, 33–100 mg L^{-1}) and starch (2 g L^{-1}) – two common components in real textile wastewater – and other nutrients into tap water. The other components of the synthetic wastewater were as follows: 0.1 g L^{-1} urea, 2 g L^{-1} KH_2PO_4, 0.099 g L^{-1} $CaCl_2$, 1.025 g L^{-1} $MgSO_4{\cdot}7H_2O$, 0.001 g L^{-1} thiamine and 1 mL L^{-1} trace elements. Stock trace elements solution was prepared by dissolving 0.125 g $CuSO_4{\cdot}5H_2O$, 0.05 g H_2MoO_4, 0.061 g $MnSO_4{\cdot}5H_2O$, 0.043 g $ZnSO_4{\cdot}7H_2O$, 0.082 g $Fe_2(SO_4)_3{\cdot}14H_2O$ in 1 L of Milli-Q water [19]. Acid Orange II (Sigma–Aldrich Co., USA) is a low molecular weight (350 g mol^{-1}) soluble, orange dye (peak absorbance at 481 nm). Granular activated carbon (GAC, F400-OS) was received from Calgon Mitsubishi Chemical Corporation, Japan. According to the manufacturer, the average particle size and pore diameter of the GAC was 1.1 mm and 2.44 nm, respectively. The GAC possessed a BET surface area, micropore volume and mesopore volume of 793 m^2 g^{-1}, 0.363 cm^3 g^{-1}, and 0.123 cm^3 g^{-1}, respectively.

2.2. Batch test description

2.2.1. Harvesting crude enzyme

Pure cultures of fungi were grown into an agglomerated mass [20] in synthetic wastewater (Section 2.1) excluding dye. Following the confirmation of secretion of the target amount of enzyme (see Section 2.5), the medium was harvested as 'crude enzyme' by filtration using a 0.45 μm cellulose acetate filter. In order to capture the effect of co-existence of other organics along with enzyme on dye degradation, enzyme solutions possessing enzymatic activity (E) and total organic carbon (TOC) of 7–98 μM min^{-1} and 62–138 mg L^{-1}, respectively, were obtained by varying the dose of starch (main contributor of TOC) and the incubation period. Accordingly, the extent of dye degradation (see Section 2.2.3) was assessed as a function of E/TOC ratio, in addition to absolute E.

2.2.2. Incubation of enzyme-preloaded GAC with dye

Two 100 mL conical flasks containing previously washed GAC (2 g) were set up. Initially, both flasks contained 70 mL Milli-Q water. The flasks were autoclaved to eliminate microbial contamination and cooled to room temperature. In the test flask, water was replaced by the same volume of crude enzyme. The contents of both flasks were then stirred (70 rpm) using a magnetic stirrer for 12 h. The respective media from the flasks were decanted. The amounts of enzyme and TOC adsorped to GAC were estimated from the difference of their respective levels in the original crude enzyme solution and the spent, decanted media. Dye solution (70 mL of 1 g L^{-1}) was added aseptically into the control (only GAC) and enzyme-preloaded GAC flasks and stirred for 1 day for complete decoloration of the media.

2.2.3. Estimation of dye degradation

At the end of the experimental period, the decolored liquid media were decanted and methanol (80 mL) was added to each flask. In order to enhance dye extraction from the activated carbon, the flasks were placed on a hot-plate stirrer to apply strong stirring (150 rpm) and intermittent heating at 1 min on/off intervals. Within a few minutes after the mixtures started boil, heating was discontinued and the methanol solution along with the extracted dye was collected by filtration using a 0.45-μm cellulose acetate filter. This sequence was repeated several times till the amount of dye extracted was negligible (as judged by the absorbance of the media at 481 nm, see Section 2.5). Under these experimental conditions, the dye extraction efficiency for GAC was 95%. Dye degradation resulting from enzymatic activity was estimated from the difference in the amount extracted from flasks containing only GAC and those containing enzyme-preloaded GAC. The degradation percentage calculated in this way was multiplied by the extraction efficiency and the resultant value was reported as percentage minimum dye degradation.

Download English Version:

https://daneshyari.com/en/article/635250

Download Persian Version:

https://daneshyari.com/article/635250

Daneshyari.com