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Visualizing Fouling Layer in Membrane Bioreactor

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The membrane fouling layer in the combined aerobic granular membrane bioreactor (AGMBR) process and the conventional MBR process was probed in this study using the multiple staining and confocal laser scanning microscope (CLSM) technique. The fouling layer built up outside and inside the membrane was principally composed of live cells embedded in a matrix of proteins and polysaccharides. The fouling layer presented outside the membrane is noted denser in structure than that inside the membrane.

Keywords CLSM; fouling; hollow-fibre membrane; membrane bioreactor

INTRODUCTION

Smith et al. (1) reported on the combined use of membranes in biological wastewater treatment. The main advantage of using the membrane bioreactor (MBR) technology over using other conventional biological processes is to produce quality water from municipal wastewater for reuse, meeting the need for saving water, particularly in regions of water shortage. Yamamoto et al. (2) developed the first submerged type MBR that are in wide use today. Membrane fouling results in flux decline and fouling increases pressure drop across the membrane (3). Sridang et al. (4) analyzed the fouling potential of a membrane utilizing different module configurations and hydrodynamic environments. Seo et al. (5) determined that the hydrophobic fraction of organic compounds fouled the membrane more than did hydrophilic fraction. Jarusutthirak et al. (6) indicate that polysaccharide colloids accounted for most fouling of UF and NF membranes. Cho et al. (7) argued that polysaccharides and related substances are the principal foulants of UF and NF membranes. Rosenberger et al. (8) indicated the impact of soluble or colloidal fractions in organic substances, particularly polysaccharides, on membrane fouling, and thereby arguing for characterizing liquid-phase compositions when monitoring the membrane process performance. Ramesh et al. (9,10) revealed the potentials of individual components in EPS to foul the MBR membrane.

Aerobic granules are considered to be a special case of biofilm composed of self-immobilized cells. The first patent was granted by Heijnen and van Loosdrecht (11). de Kreuk et al. (12) and Adav et al. (13) provided comments on the state of the art for the aerobic granulation process. Compact structured, biologically efficient aerobic sludge granules with wide diverse microbial species and excellent settling capabilities have been developed in sequencing batch reactors (SBR) (14–25). Tay et al. (26) proposed a new process, called the aerobic granular sludge membrane bioreactor (AGSBR), that combined MBR and the aerobic granule technology for wastewater treatment. The system indicated excellent membrane permeability with aerobic granules in a four months long operation compared with the conventional MBR, and it was claimed that low membrane fouling was induced by the high strength of the aerobic granules in the surface cake. Jun et al. (27) also noted enhanced filterability for the aerobic granular membrane over the conventional biofloc systems. The authors concluded that the main components for fouling are proteins and polysaccharides in water. Thanh et al. (28) utilized a baffled membrane filtration unit as the post-treatment unit of an aerobic granular sludge process. These authors noted that the soluble polysaccharides content increases with increasing organic loadings, and contributes most of the membrane resistance during the filtration of the effluent from the aerobic granular process. Wang et al. (29) developed their aerobic granular sludge membrane bioreactor (GMBR), which showed good organics removal (TOC removal $> 84.7\%$) and simultaneous nitrification and denitrification (SND) performances for synthesized wastewater.

Fast filtration with minimum irreversible membrane fouling is desired (30–37). The fouling layer develops on the membrane surface of MBR and AGMBR for raising trans-membrane pressures. For the first time Juang et al. (38) provided direct observation of an external fouling layer which occurred on a hollow-fiber membrane of the AGMBR. This work demonstrated the formation of not

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only an external but also an internal fouling layer residing inside the membrane.

EXPERIMENTAL

Reactor and Operation

Aerobic granules were cultivated with the procedures described by Adav et al. (18). Stable granules formed in three weeks. The cultivated, mature granules were washed with pure water and then stored at 8°C for 60 days in reagent bottles containing synthetic wastewater with compositions the same as those used by Adav et al. (18) combined with 250 mg l^{-1} phenol and 400 mg l^{-1} peptone.

Two laboratory-scale MBR were utilized in the present study (Fig. 1(a)). The membrane module was made of polyethylene hollow-fiber membranes of 500 μm diameter and 0.4 μm pores, provided by Mitsubishi Rayon Co. (Tokyo, Japan). The specifically prepared membrane module had a working surface of 0.03 m^2. Coarse bubbles were produced below the membrane module at a flow rate of 3 l min^{-1}.

A synthetic substrate with phenol as the sole carbon source was prepared with the following composition (gl^{-1}): pheol 2.5, NH_4Cl 1.0, K_2HPO_4 8.25, KH_2PO_4 6.75, and $MgSO_4 \cdot 7H_2O$ 0.65. The prepared substrate has the COD contribution of 3000 mgCOD l^{-1}. The substrate was continuously fed at the bottom of the MBR at 1.2 ml min^{-1}. The effluent was drawn at an on-off mode of 15 min-15 min partition.

Staining and Fluorescent Tests

To probe the structure of the fouling layers, the present work employed the confocal laser scanning microscope (CLSM, OLYMPUS BX50) equipped with an image processor (OLYMPUS FV5 PSU) and an argon laser source to stimulate the fluorescence. The granule was imaged with a 10x objective with the software FLUOVIEW version 3.0. All probes were purchased from Molecular Probes (CA, USA). All primary antibodies were purchased from Abcam (Cambridge, UK) or Invitrogen (Carlsbad, USA).

SYTO 63 (20 μM) was used to stain the nucleic acid in granule, then the stained sample was placed on the shaker table for 30 min. EPS staining is followed by the multiple-labeling immunohistochemical tests. The α-amylase antibody ab6615 from rabbit, and the tyrosinase antibody ab53338 from sheep were dipped at 1:20 dilution onto the sections and were incubated for 2 hr at 37°C. After removing residual primary antibody solution, the sections were incubated with 2 mg ml^{-1} Alexa Fluor® 568 goat anti-rabbit IgG Cy3-conjugated A11031 antibody, and 2 mg ml^{-1}Alexa Fluor® 633 goat anti-sheep IgG Cy5-conjugated A21071 antibody at 1:50 dilution for 40 min at 37°C. Finally the stained section was washed twice using the 1% v/v Tween 20 PBS solution.

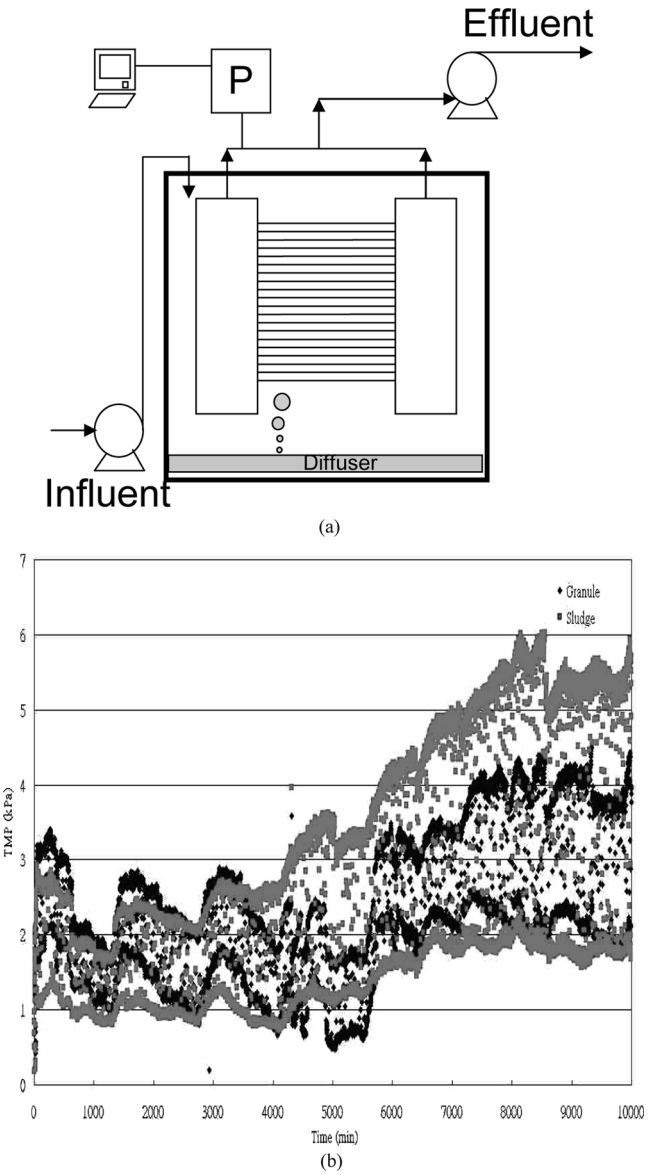

FIG. 1. (a) Schematics of the testing MBR. (b) Trans-membrane pressure of MBR and AGMBR as a function of testing time.

RESULTS AND DISCUSSION

Reactor Performance

The COD and TOC data in the effluents and/or supernatants for AGMBR and MBR tests revealed that both reactors performed satisfactorily over the testing time. The removal of COD and TOC was mostly >95%.

Figure 1(b) shows the time courses of trans-membrane pressures (TMP) for AGMBR and MBR tests. The AGMBR was seeded with the cultivated granules while the MBR was seeded with activated sludge from a local wastewater treatment plant. The TMP for MBR started to increase at $t > 4000$ min, with the peak pressure being raised to about 4 kPa at $t > 6000$ min. Regular backwashing

could almost completely restore the membrane flux. For AGMBR, the TMP could keep low until $t = 5700$ min. Afterwards the peak TMP rapidly increased to >5 kPa. Hence, the AGMBR delays the occurrence of membrane fouling when compared with the MBR tests.

Fouling Layer Visualization

Figure 2(a) presents the fluorescent staining results of the single fiber positioned close at the central regime of the AGMBR at the end of test. The fiber was physically washed. The surface of the membrane was relatively clean compared with the mass of "sludging" noted in Juang et al. (38). Conversely, an internal biofilm was noted. The imaging results for MBR revealed the presence of similar internal biofilms (figure not shown for brevity). Parallel tests demonstrated that these internal biofilm could not be effectively removed by either physical or chemical cleaning.

Figures 2(b)–(d) presents the magnified, fluorescent staining results of the external and internal biofilms noted in Fig. 2(a). A very heterogeneous layer of thickness of 30 μm was revealed. Redundant cells were noticeable embedding in a matrix, with a layer of tyrosinase surrounding the said cells. The internal biofilm was looser in packing and thinner than that for the external biofilm layer. The presence of tyrosinase activity correlated with the high phenol removal detected in the present MBR system. Additionally, the quantity of α-amylase was negligibly detected. Restated, the degradation of polysaccharides was not activated in the studied reactors.

Penetration of live cells through the membrane should contribute to the occurrence of the internal biofilm layer. The AGMBR retains most live cells with the biological granules in suspension, thereby providing less chance for cell penetration. Such an observation may correlate with the delayed occurrence of TMP rise as noted in Fig. 1.

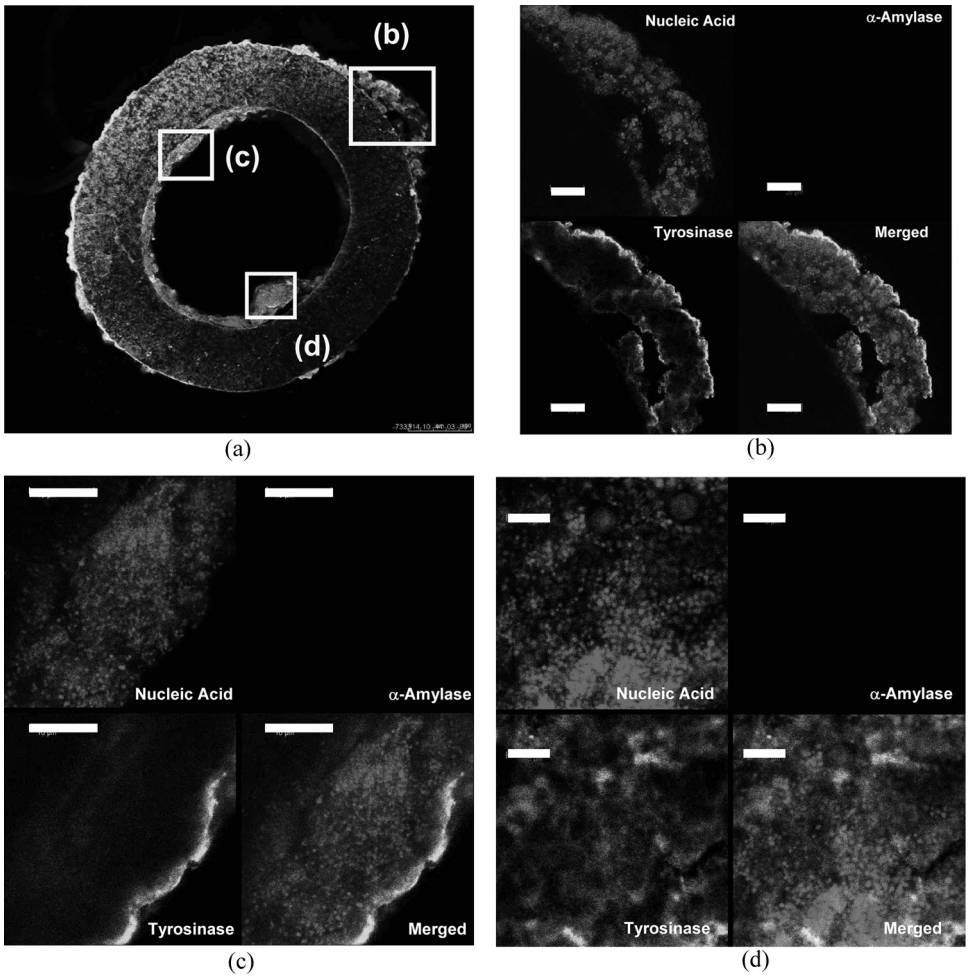

FIG. 2. The CLSM images for the fouling layers on single hollow-fiber membrane in AGMBR. (a) CLSM image of the sectioned membrane fibre. (b)–(d) are the magnified images of those marked in (a). Bar = 10 μm. Envelope layers in (b) and (c) and cloud-like in (d): tyrosinase; light patches in background: alpha-amylase; dots: cells. Bottom and right: combined image.

CONCLUSIONS

The AGMBR and MBR were tested with phenol-containing synthetic wastewater with TMP data probed over time. Both external and internal biofilms were developed on the membranes. The CLSM images on a single fiber revealed the presence of very heterogeneous layers of thickness of 30 μm with redundant cells. These cells were embedded in a matrix, with a layer of tyrosinase surrounding them. The internal biofilm was looser in packing and thinner than that for the external biofilm layer. Very likely both external and internal biofilm developments determined the membrane permeability in MBR operation.

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