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Effects of step-feeding and internal recycling on nitrogen removal in ceramic membrane bioreactors, and their hydraulic backwashing characteristics

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ABSTRACT

Both lab-scale (50 L/day) and pilot-scale (10 m^3/day) ceramic membrane bioreactors (CMBRs), equipped with two additional implements (step-feeding and internal recycling), have been tested for the removal of nitrogen and organic matter from municipal wastewater. During the 180 days of operation, 0.5Q of step-feeding increased the removal rate of total nitrogen by 46.2% in the lab-scale CMBR, whereas a 59.0% increase was observed with the additional 3Q internal recycling in the pilot-scale CMBR. This was presumably due to the supply of insufficient carbon source as an electron donor in the denitrification stage. The ceramic membrane also exhibited high rigidity sufficient for periodic backwashing using the highly-pressurized permeate. A constant flux test with 30 s of backwashing after every 9.5 min of filtration successfully removed foulants from the pores of the ceramic membrane, and instantaneously restored the trans-membrane pressure (TMP). During the test, the TMP in case of without the hydraulic backwashing steadily increased (20–80 kPa) for 40 min, whereas it took more than two times to reach the same TMP in the case of with the hydraulic backwashing, which eventually doubled the time span between required chemical washings.

1. Introduction

Ceramics, made of titanium–aluminum oxides and silicon carbides, have been used in various industries because of their stability, high melting point (more than 3000 °C) and hardness (6 GPa) [1]. Because of their superior characteristics of chemical resistance, thermal and mechanically stability, and high permeability [2], ceramic membranes have also been applied to broad fields such as gas separation [3], desalination [4], and fuel cell [5]. An oxygen-ion-conducting ceramic membrane is used for oxygen separation in various applications involving the metallurgical industry [6], cures for chronic lung diseases by oxygen separation [7], and also production of valuable syngas ($CO + H_2$) from methane [3]. In addition, the used of ceramic membranes has broadened to include not only gas separation but also to a whole new range of industrial uses including wastewater treatment [8].

There have been extensive research efforts to remove organic matter and nutrients from wastewater using various types of biological treatment removal (BNR) processes (e.g., pre-denitrification (A/O), anaerobic/anoxic/oxic (A^2/O), and the University of Cape Town (UCT) process) [9]. Recent studies have proposed use of a biological process combined with use of a membrane, called a membrane bioreactor (MBR) [10]. By adoption of MBR, settling basins for solid–liquid separation after biological treatment have been replaced by membranes, and the new process can be operated regardless of the condition of the activated sludge [11].

Sometimes the organic carbon available is insufficient for complete removal of nutrients in enhanced biological wastewater treatment processes [12]. In addition, microorganisms such as denitrifiers and phosphorus accumulating organisms (PAOs) compete for organic carbon for denitrification and phosphorous release, while at the same time enhancing their cell growth and maintaining their community [13]. There have been several studies aimed at resolving this shortage problem. Baeza et al. [14] applied various ratios of internal recycling of the influent to increase the nitrogen removal efficacy for the A^2/O process, finding that the nitrogen

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removal efficiency was increased by nearly 12.3% when the internal recycling ratio was increased to five. Step-feeding is another alternative technique for enhancing nitrogen removal by introducing an organic carbon source into the influent of each tank [12].

As the material science and technology of membranes has developed, various types of polymer membranes were applied to the MBR process. Pore sizes appropriate for microfiltration and ultrafiltration were the most common, and various types of membrane modules of flat sheets or hollow fibers have been commercially available for several decades. However, there is a chronic problem with polymer membranes. Recent studies aimed at enhancing the resistance of polymer membrane enabled acid/base washing to reduce fouling. However, there is still the big problem of long term maintenance, which requires high thermal, mechanical and chemical stability [15]. Physical and chemical washing is mandatory in order to reduce biological fouling of membranes, and to restore the flux by reducing the trans-membrane pressure (TMP). Membrane washings, however, can damage the polymer surfaces and membrane materials over the long-term. Additionally, chemicals used in washing can result in secondary pollution in the case of spills into the effluent [16]. For this reason, more efficient methods of physical washing are required to minimize the mandatory chemical washing. Among them, membrane backwashing is one effective method that can restore the membrane flux to its initial level, whereas simple water rinsing cannot remove accumulated deposits from pore spaces [17]. Ceramic membranes, as mentioned above, have enough rigidity to endure physical backwashing owing to its mechanical durability, whereas only limited types (e.g., tubular type) of polymer membrane can be used with backwashing. There have been several studies using ceramics as membrane materials for municipal wastewater treatment before [18–20]. Most of these; however, were limited to merely evaluating the performance of the ceramic membranes for organic matter removal in small, lab-scale experiments.

In this study, two different scales of ceramic membrane bioreactors (CMBRs) were tested to investigate the effect of both step-feeding and internal recycling on the efficacy for removal of nitrogen and organic matter, using both simulated and real wastewater. First, 50 L/day of lab-scale CMBR was tested using simulated wastewater, varying the operation modes (internal recycle and step-feeding). The results were extended to a 10 m^3/day pilot-scale CMBR using real wastewater accounting for the size effect. Additionally, the influence of backwashing on TMP during CMBR operation, was also investigated by comparing operation modes with and without hydraulic backwashing.

2. Materials and methods

2.1. Preparation of ceramic membrane

The ceramic membranes used in both the lab-scale and pilot-scale CMBRs, were supplied from Meidensha Corporation, Japan, and were made of alumina. The membrane type was flat-sheet, which was operated using the out-in filtration method. Table 1 shows the specifications of the membrane module used in this study. For the lab-scale test, the module was composed of three flat-sheets, each 93 × 93 × 7 mm. Meanwhile, that used in the Pilot-scale plant consisted of two membrane units, each 780 × 1740 × 420 mm, and with 7.7 m^2 of effective area. Each was composed of seven modules, and each module (480 × 160 × 230 mm) was composed of 12 flat sheets. Prior to the experiment, the surface of the ceramic membrane is observed by using field emission gun–scanning electron microscopy (FEG–SEM) (Inspect F50; FEI, Eindhoven, The Netherlands) operated at 10–20 kV, equipped with an energy dispersive spectroscopy (EDS) analyzer. All samples were dried in a vacuum oven for 6 h at 60 °C before analysis.

Table 1
Specification of the ceramic membrane modules used in the lab-scale and pilot-scale CMBRs.

Item	Specification
Pore size (μm)	0.06
Durability	Maximum TMP: 100 kPa
	Maximum temperature: 40 °C
	pH range: 1–10
Materials	Alumina (Al_2O_3)
Size (mm)	93 W × 93 H × 7 T (lab)
	480 W × 160 H × 230 T (pilot)
Effective area (m^2)	1.1 (lab)/7.7 (pilot)

2.2. Lab-scale CMBR operation

A schematic diagram of the CMBR used in this study is presented in Fig. 1. The lab-scale reactor consisted of a pre-denitrification tank, primary intermittent aeration tank, secondary intermittent aeration tank, and filtration tank, in which a ceramic membrane with effective area of 0.052 m^2 was submerged. The capacity of the reactor was 50 L/day, and the specification of each tank is shown in Table 2.

The operation modes of the lab-scale CMBR (mode 1 and mode 2) are described in Table 3. Air was supplied by blower, and the air rate controlled with a flow regulator at 3–4 mg DO/L. Aeration at the both intermittent aeration tanks was alternated every 45 min by using a programmable logic controller (PLC). Filtration and hydraulic backwashing were alternated every 9.5 min, and 30 s, respectively. If the filtration time is too long, there might be a build-up of irreversible fouling, whereas if the time is too short, unnecessary amount of permeate will be wasted for the backwashing [21]. The filtration and backwashing periods in this study were determined basically from the batch flux test maintaining constant flux (20 LMH). After the 10 min of filtration, TMP already increased up to 40 kPa, and reached 50 kPa in 20 min, a maximum allowable pressure that requires intensive physical and/or chemical cleaning. A vacuum gauge was installed between the membrane module and the filtration pump for monitoring the TMP. To investigate the effect of step-feeding on nitrogen removal, modes 1 and 2 in the lab-scale CMBR were conducted as presented in Table 3. Both modes had internal recycling from the secondary intermittent aeration tank to the primary intermittent aeration tank, at three times the inflow rate (3Q). The difference between mode 2 and mode 1 was the presence of step-feeding to the pre-denitrification tank and primary intermittent aeration tank (0.5Q/0.5Q), respectively.

Several studies have focused on the step feeding and internal recycling in BNR process, and the optimum ratio fell in the range of 0.2Q–0.5Q for step feeding and 2Q–5Q for internal recycling respectively [14,22] although these primarily depend on the composition of influent (i.e., C/N ratio). Several different ratios was tested using lab-scale CMBR, and the best combination was found to be 3Q for internal recycling, and 0.5Q for step feeding. Sludge wasting was manually carried out on a daily basis (0.17–0.25 L/day) at the filtration tank considering the mixed liquor suspended solids (MLSS) concentration, which provides the solids retention time (SRT) of ca. 50 days. The composition of the synthetic wastewater used in the lab-scale CMBR is listed in Table 4.

2.3. Pilot-scale CMBR operation

The 10 m^3/day pilot-scale CMBR was installed at the Osan Sewage Treatment Plant (STP) in Korea. Schemes for the overall process, and for the ceramic membrane modules and units, are shown in Figs. 1 and 2, respectively. The specifications of each tank in the pilot-scale CMBR were also shown in Table 2. The pilot-scale CMBR was seeded with activated sludge from a return line in the