



# Influence of $Mg^{2+}$ catalyzed granular sludge on flux sustainability in a sequencing batch membrane bioreactor system

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## HIGHLIGHTS

- Granular sludge with controlled  $Mg^{2+}$  augmentation maintained the high flux rates.
- Membrane permeability of granular sludge was 6-fold higher than the activated sludge.
- 50% membrane fouling reduced by aerobic granules and low SMP contents.
- FTIR & SEM revealed proteins, polysaccharides and fine particles as major foulants.

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## ABSTRACT

The present study was designed to achieve steady state high membrane flux values by controlling the organic and inorganic foulants during MBR operation. Three granular sludge sequencing batch membrane bioreactors, namely, R-1, R-2 and R-3, were run at a constant flux of 40 LMH for 90 days. R-1 was operated without the addition of  $Mg^{2+}$ , R-2 with a continuous dosage of 50 mg/L  $Mg^{2+}$ , whereas R-3 was run with stoichiometric amounts of  $Mg^{2+}$  (18–22 mg/L) in correlation with SMP contents. The particle sizes of aerobic granules were approximately 725 and 600  $\mu m$  in R-2 and R-3, respectively, compared with 250  $\mu m$  in R-1. The ratios of EPS proteins/polysaccharides in R-1, R-2 and R-3 were 1.63, 3.90 and 3.76, respectively, whereas the SMP concentrations in R-1, R-2 and R-3 were 40, 5 and 5 mg/L, respectively. The results highlighted that in R-3, the controlled addition of  $Mg^{2+}$  along with the emergence of aerobic granules tremendously increased the membrane permeability, which was approximately 6 and 3 times higher than R-1 and R-2, respectively. In R-3, a molar ratio of 1:2 between  $Mg^{2+}$  and SMP was found to be optimal for the successful sustainability of high flux values during the long term membrane treatment. FTIR, SEM and ICP spectroscopic investigations revealed that the deposition of fine sludge flocs and high amounts of proteins and polysaccharides on the membrane surface in R-1 and  $Mg^{2+}$  in R-2 were mainly responsible for their low permeability.

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**Abbreviations:** EPS, extracellular polymeric substances; FTIR, Fourier transform infrared; SEM, scanning electron microscopy; ICP, inductively coupled plasma; MBR, membrane bioreactor; SMP, soluble microbial products; SBR, sequencing batch reactor; F/M, food/microorganism ratio; GSMR, granular sludge sequencing batch membrane bioreactor; TMP, transmembrane pressure; COD, chemical oxygen demand; PVDF, polyvinylidene difluoride; SVI, sludge volume index; MLSS, mixed liquor suspended solids; TN, total nitrogen; TP, total phosphorous; SS, suspended solids.

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## 1. Introduction

The membrane bioreactor (MBR) process has suddenly become an attractive choice for treating wastewater, as evidenced by their rapid installation in recent years. The significance in using MBR for wastewater treatment is the achievement of superior effluent quality, compared to the conventional activated sludge systems. In addition, MBR provides a complete control of solids and a smaller footprint [1,2]. However, the flux decline due to membrane fouling is the biggest challenge being faced by water technologists these days, as it decreases the membrane efficiency, resulting in increased operational and maintenance costs [2,3]. The organic

and inorganic foulants in soluble, colloidal and particulate form not only deposit on the membrane surface but also inside the membrane pores [4]. This causes severe blocking of pores, and the membrane needs to be cleaned chemically, which itself is a costly and hazardous process [1,2].

To control this membrane fouling, various modifications in the design of reactors, membrane structure and operational conditions have been made for the smooth running of MBRs. Studies have been performed by coupling the membrane with a sequencing batch reactor (SBR) to achieve long term steady flux rates [5,6]. The SBR system is used to synthesize the granular sludge of much higher particle size compared with activated sludge flocs [7,8]. The aerobic granules thus synthesized tremendously improve sludge characteristics such as enhanced settleability, dewaterability and the ability to withstand at high organic loadings [7,9]. Tay et al. [10] reported that at constant flux tests, the loss of membrane permeability in MBR containing granular sludge was 21-fold lower, compared with MBR containing flocculant sludge. Similarly, some other researchers also found that the MSBR (membrane sequencing batch reactor) system effectively controlled the membrane fouling than that of MBR due to the introduction of aerobic granules in the former [6,11]. Feng et al. [12] while comparing the fouling tendency of floc particles and granular sludge, highlighted that the presence of high moisture contents and negative surface charge on the floc sludge were the main causes for their low permeate flux values than the aerobic granular sludge. However, the aerobic granules are cultivated at high organic loading rates, and there is a potential risk that it might induce organic fouling [13]. In fact, at high food/microorganisms (F/M) ratios, the microbial activities are increased, resulting in the production of higher amounts of extracellular polymeric substances (EPS). These biopolymers are present either as a part of granular sludge which are referred as EPS or in the soluble form which are referred as soluble microbial products (SMP) [14]. It has been stated that SMP is a major source of membrane fouling that severely affects membrane efficiency [15–17].

Some researchers added up to 830 and 96 mg/L of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , respectively, to enhance the membrane permeability in an MBR system containing activated sludge flocs [18,19]. They reported that the organic fouling, particularly due to soluble biopolymers, was effectively reduced in the presence of bivalent metals. The EPS contained an abundant amount of anionic functional groups and are precipitated in the wastewater by interacting with these metallic cations. However, the continuous addition of high metal contents induces inorganic fouling due to precipitation reactions which along with the organic matter favors the formation of a gel layer on the membrane surface, thus deteriorating the membrane permeability [20]. Moreover, the addition of high quantities of metal increases the operational cost, thus making it very hard to utilize on a commercial scale.

By considering these problems, during the current studies, a hybrid process was developed in which the aerobic granules cultivated by  $\text{Mg}^{2+}$  addition were coupled with a hollow fiber membrane bioreactor (GSMBR) system. The main objective was to achieve stable high membrane flux values by controlling the membrane fouling due to soluble species, floc particles and metal precipitates. In this regard, two operating strategies were followed: (1) one GSMBR was run with aerobic granules cultivated with continuous addition of 50 mg/L  $\text{Mg}^{2+}$  in the influent wastewater, and (2) the other was run with granular sludge synthesized by feeding 18–22 mg/L  $\text{Mg}^{2+}$  in a molar ratio of 1:2 with SMP contents. The process performances of both GSMBRs were compared by running a control reactor without  $\text{Mg}^{2+}$  addition and containing the activated sludge flocs.

## 2. Materials and methods

### 2.1. Operation of membrane bioreactors

Three identical laboratory-scale GSMBRs, namely R-1, R-2 and R-3, were run for 90 days (a schematic diagram of the GSMBR is shown in Fig. 1). The total volume of each reactor was 5.60 L, with a workable volume of 5 L. R-1 served as a control reactor and operated without the addition of  $\text{Mg}^{2+}$  in the influent wastewater. The COD in this reactor was adjusted at 200 mg/L (similar to real municipal wastewater in the activated sludge treatment) by adding glucose. In R-2, the influent COD was 500 mg/L, and the  $\text{Mg}^{2+}$  augmentation was 50 mg/L using  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  salt. In the case of R-3, COD was again 500 mg/L, whereas  $\text{Mg}^{2+}$  was injected in a theoretical 1:2 ratio with soluble contents by considering that one divalent cation can bind with two negatively charged polymeric species.  $\text{Mg}^{2+}$  dosages were decided by randomly measuring the SMP contents at the end of the cycle once per day. Following this criteria, 18–22 mg/L  $\text{Mg}^{2+}$  was added in R-3 from days 10 through 90 of the experiment. The organic loading rate in terms of food-to-microorganisms (F/M) ratio in R-1, R-2 and R-3 was 0.20, 0.50 and 0.50 kg COD/kg MLSS-d, respectively. It has been reported that the granular sludge is synthesized at high loading rates while at lower COD loading rates the sludge normally exists as floc particles [7]. Thus the loading rate was fixed at a lower value in R-1 than R-2 and R-3, so that the filtration characteristics of the R-1 floc particles can be compared with the aerobic granules of R-2 and R-3. The temperature of the reactors was adjusted to 20 °C by wrapping them with thermally controlled heating bands. The reactors were operated on a 6 cycles/day basis (cycle duration = 4 h). During the first 10 days (before the appearance of the granules), each cycle consisted of 10 min filling, 195 min aeration, 30 min settling and 5 min effluent withdrawal. The influent injected from the top, and the effluent discharged from the center of each reactor through the solenoid valve. From day 11 (after the appearance of aerobic granules), three hollow fiber membrane modules made up of PVDF (polyvinylidene difluoride) were taken and immersed in each reactor for the removal of treated water using the suction pump. In this stage, the settling and effluent drainage periods were eliminated, and the revised experimental conditions were 10 min filling, 228 min aeration, and 2 min idle time. Each membrane module had an effective surface area of 0.04 m<sup>2</sup>, a pore size of 0.45 µm, and the filtration was carried out at a constant flux of 40 LMH (liter/m<sup>2</sup> h). The membrane was run only during the aeration mode after 1 h of the influent injection; intermittently in on/off cycles of 8 and 3 min, respectively, to control the fouling rate. The water turbulence during the aeration

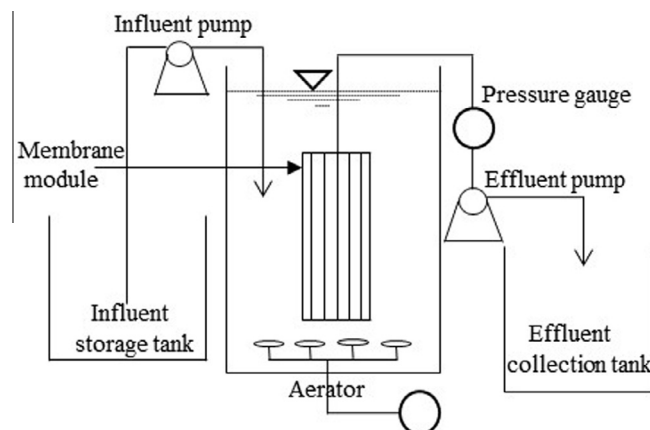


Fig. 1. A schematic diagram of the GSMBR.

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