



# Determination of fouling-related critical flux in self-forming dynamic membrane bioreactors: Interference of membrane compressibility

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## ABSTRACT

A new measurement protocol was proposed for appropriate determination of the fouling-related critical flux of highly compressible self-forming dynamic membranes (SFDMs) developed on coarse-pore supporting materials. Intermittent relaxation periods and parallel clean water filtration tests were incorporated into the common transmembrane pressure (TMP) step method to minimize the interference of SFDM compressibility on fouling-related critical flux determination and to better understand the filtration characteristic of SFDMs. The average fouling-related critical flux of SFDMs determined by the new measurement protocol turned out to be significantly higher than that obtained with the common measurement protocol. This is attributed partly to the exclusion of the flux reduction associated with SFDM compression by conducting the parallel clean water filtration tests, and partly to the minimization of the effect of “continuous” compression on the structure and fouling propensity of SFDMs by integrating the intermittent relaxation periods. The intermittent relaxation periods also prevented the occurrence of irreversible fouling and steady compression of SFDMs, embodied by the relatively smaller and faster disappeared hysteresis observed in the new measurement protocol. As one way to control SFDM compression, intermittent effluent production mode appears to be a potentially promising strategy for further optimization of SFDMBR operation. Stepwise increment of relaxation time and proper selection of higher initial TMP turned out to be the options recommended for reducing the time consumption of the new measurement protocol.

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## 1. Introduction

Great efforts were dedicated to the R&D of new configurations and modifications of membrane bioreactors (MBRs) as recently reviewed by Meng et al. [1]. One innovative and promising technology is termed self-forming dynamic membrane bioreactors (SFDMBRs) [2–7]. In SFDMBRs, the filtration modules are made of coarse-pore materials (e.g. non-woven fabric, nylon mesh, and stainless steel mesh) instead of microfiltration (MF)/ultrafiltration (UF) membranes employed in conventional MBRs [8–11]. Though the coarse-pore materials themselves cannot adequately fulfill the solid-liquid separation due to their over-large pore size, MF/UF equivalent separation efficiency can be achieved by actively utilize the self-forming dynamic membrane (SFDM), i.e. the initial “desired” sludge layer developed on the coarse-pore supporting materials during the initial period of sludge filtration.

SFDMBRs are more affordable than MBRs due partly to the substitution of MF/UF membranes by less expensive coarse-pore materials, and partly to the lower energy consumption of SFDM filtration [4,9,10]. This makes SFDMBRs a highly potential alternative of MBRs especially in developing countries where the widespread application of MBRs remains an economic challenge. However, similar as MF/UF membrane fouling in MBRs, SFDM fouling arising from the subsequent “undesired” sludge deposition/adsorption after SFDM formation results in increasing filtration resistance and operating cost, which greatly deteriorates SFDMBR performance. Understanding and mitigation of SFDM fouling are therefore of fundamental importance to the implementation and optimization of SFDMBRs.

Membrane fouling in MBRs can be effectively characterized and mitigated via critical flux determination and sub-critical flux operation, respectively [12–15]. As one of the most important concepts in membrane fouling studies, critical flux was initially proposed in 1995 [16–18] and defined, in principle, as the flux below which fouling does not occur (strong form) or fouling is independent of solvent transfer (weak form). Critical flux has since been investigated by numerous researchers, reviewed in-depth by Bacchin et al. [19], for better understanding of membrane fouling

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characteristics. Though critical flux was extensively investigated in MBR fouling [12–15,20–22], no special efforts have yet been devoted to understanding the fouling-related (i.e. excluding the interference of compression as discussed later) critical flux of SFDMs. In fact, the basic measurement protocol for fouling-related critical flux determination of SFDMs has not been established, probably because of the relative novelty and distinctive characteristics of SFDMs.

The critical flux of MF/UF membranes is commonly determined by continuous flux or transmembrane pressure (TMP) step methods, where membrane fouling is indicated and evaluated by the increase of filtration resistance at each step [19,23]. These methods, however, cannot be directly applied for proper measurement of the fouling-related critical flux of SFDMs because of the potentially significant interference of SFDM compression. Unlike MF/UF membranes of rigid structure, SFDMs are indeed the initial sludge layers deposited/adsorbed on/in coarse-pore supporting materials and have relatively loose structure with high compressibility. They may thus experience “continuous” compression during common critical flux determination trials. The term “continuous” compression herein was particularly referred to the continuously stepwise increasing compression underwent by SFDMs along with the continuously stepwise increase of flux or TMP in the course of critical flux measurement. This manner of compression should therefore be minimized as it was not the case in practical SFDMBR operation wherein the designed flux/TMP was not set via step-by-step increase. The “continuous” compression of SFDMs taking place during critical flux measurement would stepwisely alter the original structure and fouling propensity of SFDMs, making filtration with the same SFDM at different steps impossible.

In addition, SFDM compression occurring during critical flux measurement would result in significant increase of filtration resistance regardless of fouling. Fouling-related critical flux of highly compressible SFDMs thus cannot be determined by currently available measurement protocols, which cannot effectively evaluate and then remove the interference of SFDM compression. In other words, the fouling behaviors and moreover the unique filtration characteristic of highly compressible SFDMs cannot be elucidated without taking into account the interference of SFDM compression.

The present study attempts to develop a theoretically sound new measurement protocol for more accurate determination of the fouling-related critical flux of SFDMs. Endeavors focus on minimizing the interference of “continuous” SFDM compression on fouling-related critical flux determination by (i) adding intermittent relaxation periods to fast loosen the SFDM structure compressed in the previous step back to its original state, and (ii) conducting parallel clean water filtration tests to evaluate and then remove the contribution of SFDM compression to the increase of filtration resistance. A series of comparative tests were carried out in a specially designed isolable multi-compartment SFDMBR for rigorous evaluation and subsequent optimization of the proposed measurement protocol. The acquired results would provide valuable insights into the concept of fouling-related critical flux of highly compressible SFDMs, and further deepen the understanding of the filtration characteristic in SFDMBRs.

## 2. SFDM compressibility

The compressibility of a cake layer is characterized by its increasing specific cake resistance with increasing TMP. Though it is widely reported that the cake layer formed on MF membrane surface during filtration of microbial suspensions exhibits certain degree of compressibility [24], few attempts have yet been made to understand the compressibility of SFDMs, a special type of cake layer developed on coarse-pore supporting

materials. The compressibility index of SFDMs was estimated to be 0.64 (Fig. S1 in Supplementary information) according to the steady-state method detailed described in Supplementary information, which proves the highly compressible nature of SFDMs. Kiso et al. [8] also reported that sludge layer formed on nylon mesh (100–500  $\mu\text{m}$  pore size) can be easily compressed even under very low pressures, though they did not quantify the compressibility index.

Due to the “continuous” compression as flux or TMP stepwisely increased in the course of critical flux determination, the porous structure of the highly compressible SFDM cannot be maintained the same as its original state if no relaxation periods were applied. The porosity of the SFDM would instead decrease at the subsequent higher flux/TMP step. The decreased SFDM porosity would result in higher fouling propensity particularly to small size sludge components that can be more readily captured by the narrowed pores, and meanwhile lead to higher specific cake resistance according to Carman–Kozeny equation. This means that the SFDMs tested at different flux/TMP steps below critical flux can no longer be expected to be the same with each other due to the “continuous” compression. In other words, critical flux measurement was not done with one SFDM like the case of MF/UF membranes but rather with different SFDMs having increasing more compact porous structure and higher fouling propensity as the measurement course proceeded. This would result in the determination of misleading fouling-related critical flux values.

## 3. Materials and methods

### 3.1. Experimental setup and operating conditions

A lab-scale bioreactor consisting of four identical isolable compartments (Fig. 1), divided by either short or long removable separation plates, was specially designed for proper evaluation of the fouling-related critical flux of SFDMs. The short removable separation plates were applied to optimize aeration effect similar as the common practice in MBR operations, which however were subsequently replaced by the long ones after SFDM formation for complete isolation of the four compartments.

Each compartment (working volume of 6L) contained a submerged plate-and-frame SFDM module made of polyester non-woven fabrics (0.20 kg/m<sup>2</sup>) with an effective filtration area of 0.03 m<sup>2</sup>. Aeration was done through the air diffuser installed right beneath each SFDM module for oxygen supply and activated sludge mixing. The aeration rate was controlled at  $0.6 \pm 0.03 \text{ m}^3/\text{h}$  throughout the experimental period. A mixed liquor outlet was set up beside the effluent outlet of each compartment. It served particularly for sludge discharge during the intermittent relaxation periods in the proposed measurement protocol.

The SFDMBR was operated at constant TMP mode. Due to the low filtration resistance of SFDMs, the required TMP can be fully provided by the hydraulic head difference ( $\Delta h$ ) between the bioreactor and the effluent outlet. Effluent pumps can thus be discarded in SFDMBR operation. Synthetic wastewater was supplied into the SFDMBR where the water level was automatically controlled by the water level sensor and able to be stepwisely increased/decreased according to the experimental requirements. The composition of the synthetic wastewater is listed in Table S1 in Supplementary information. The activated sludge was taken from another aerobic SFDMBR (working volume of 120 L) fed with the same type of synthetic wastewater. The decrease of effluent flux ( $J$ ) was continuously monitored at each TMP step using a measuring cylinder at preset time intervals. Effluent flux values were corrected to 20 °C

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