DETERMINING CRITICAL FLUX AND COLLOID/SMP FOULING IN A SUBMERGED ANAEROBIC MEMBRANE BIOREACTOR

ZHANG Q.[1], SINGH S.[1] and STUCKEY D.C.[1,2]

[1]Advanced Environmental Biotechnology Center (AEBC), Nanyang Technological University, Singapore, [2]Chemical Engineering Department, Imperial College London, UK, Room 516, Department of Chemical Engineering, Imperial College London, London SW7 2AZ
Email: DStuckey@ntu.edu.sg, d.stuckey@imperial.ac.uk)

ABSTRACT

A lab-scale submerged anaerobic membrane bioreactor (SAMBR) was operated with a synthetic feed of 500 mgCOD/L to determine the "critical flux" where the colloids and SMPs deposit on the membrane and increase the Transmembrane Pressure (TMP) above zero. Three different experimental strategies were employed using datalogged transmembrane pressure (TMP) data, with every constant flux step lasting for 20 min, 24 hours and 3-4 days to determine the "critical flux". The TMP profile indicated that the pragmatic critical flux of the SAMBR was 24 LMH. The colloids and SMPs were separated by filtering the supernatant through the 1 µm and 0.45 µm filters, and quantified by measuring the COD, protein and polysaccharide concentrations. The size exclusion chromatography (SEC) analysis by HPLC demonstrated the molecular weight (MW) distribution of SMPs. SMPs accounted for 46% of the overall organics of the supernatant prepared from solubilising the membrane foulants and the SMPs with a MW bigger than the 1,522 kDa were retained on the membrane, which contributed the most to membrane fouling. The colloids were present by a large percentage in the short-term operation of the reactor, but less in the long-term operation. This study provides crucial information to optimize the operation of the SAMBR, and elucidate the roles of the SMPs and colloids in membrane fouling above the critical flux.

Keywords: Anaerobic membrane bioreactor; critical flux; soluble microbial products; colloids; fouling layer

1. Introduction

Anaerobic membrane bioreactors have attracted worldwide interest in the past decade at both the academic and commercial levels due to their inherent advantages over aerobic treatment systems (Kang *et al*. 2002, Stuckey 2012). However, one of their major limitations is the reduction in flux caused by membrane fouling. Previous researchers have reported that soluble microbial products (SMPs) were critical in membrane fouling, but their composition was still unclear (Okamura *et al*. 2009). In addition, suspended colloids in the reactor with a range of different sizes had different behaviours such as aggregation or clogging when fouling the membrane (Choo and Lee 1996), but this phenomenon is still poorly understood. Therefore, the aim of this study was to examine different methods for determining the critical flux of a membrane reactor, and analysing the molecular weight distribution of the SMPs and the colloid concentrations on the membrane surface to gain a deeper understanding of their roles in membrane fouling so that reactor operation could be optimized, i.e. maximise COD removal and flux.

2. Materials and methods

A lab-scale submerged membrane bioreactor (SAMBR) was operated at a temperature of 35 °C, an HRT of 6 hours and high SRT (>150d). The sludge concentration was 3.5 - 4.5 gTSS/L, and the reactor was fed continuously with a synthetic soluble feed of 500 mgCOD/L. The biogas was circulated by a diaphragm pump with a gas flow rate of 5 litres per minute (LPM) under the membrane to scour it. Three operational strategies were employed to determine the critical flux of the SAMBR: (1) The TMP was datalogged over 20 mins with constant fluxes of 5, 10, 15, 20,

22, 24, 26, 28, 30, 33, 36 LMH, and then the flux was gradually decreased over time to 5 LMH to determine any hysteresis effects; (2) The TMP was datalogged over 24 hours at each flux between 15 and 30 LMH, and then reduced again to 5 LMH; (3) long term operation with each flux constant for 3 – 4 days. In strategy 2, all the foulants covering the entire membrane were dissolved in mineral water, centrifuged at 7,500 rpm for 10 mins, and the supernatants filtered through 1 µm and 0.45 µm membranes to quantify both colloids and SMPs by measuring the COD ($K_2Cr_2O_7$), protein (BCA standard kits) and polysaccharide (phenol-sulphuric acid method) concentrations. The size distribution of the SMPs was analysed using High Performance Liquid Chromatography (HPLC-Shimadzu) with a Polygel size exclusion column (Agilent, E*clipse Plus C18* columns). In strategy 3, an area of 5 x 5 cm of the foulants on the membrane were scraped off and dissolved in phosphate buffered saline (PBS) solution and analysed using a TOC-TN analyzer.

3. Results and discussion

3.1. Determination of critical flux in the SAMBR

A chlorinated polyester membrane (Kubota- surface area 0.116 m^2, nominal pore size of 0.2 µm) initially cleaned with hypochlorite was put in the SAMBR. Initially the SAMBR was operated at 5 LMH, and the flux climbed rapidly from 1.3 to 2.8 KPa over 10 minutes before falling back to its initial value (Fig 1a).

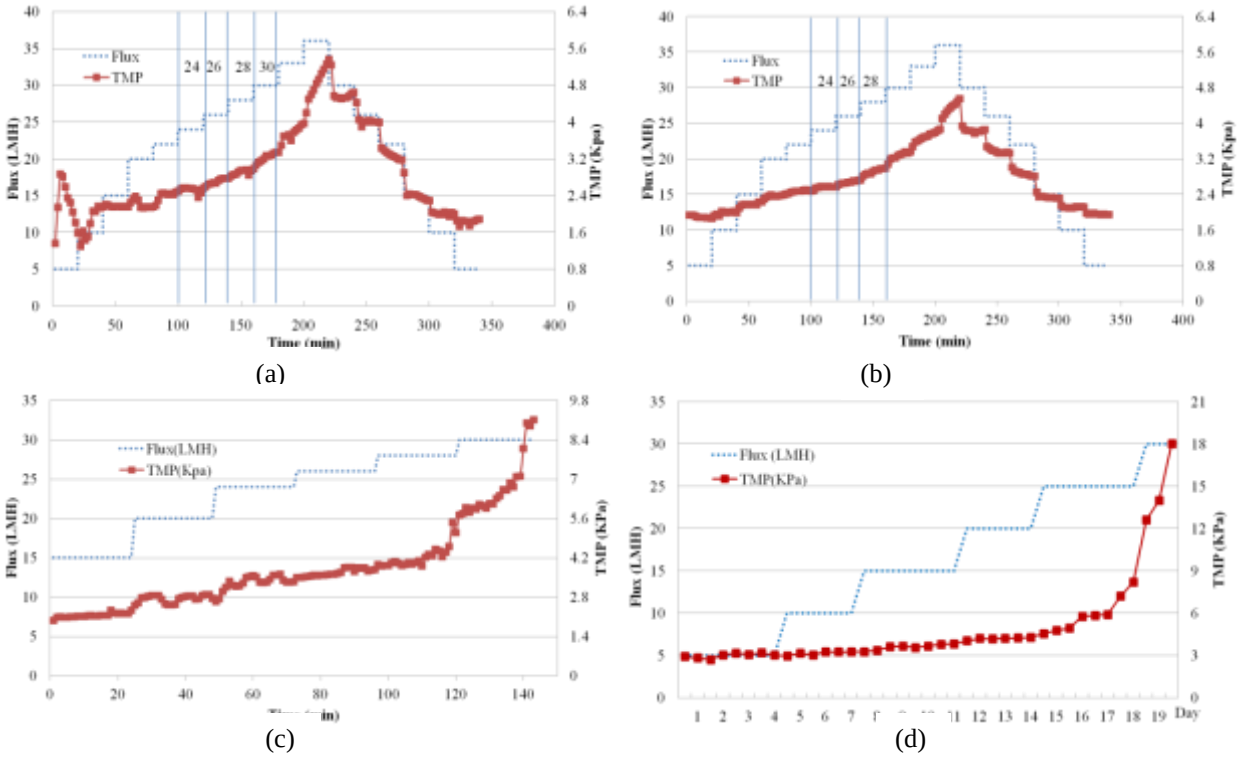


Figure 1: TMP profile with changes in flux in the SAMBR (a) start with the chemically clean membrane Strategy 1; (b) start with the fouled but relaxed membrane in strategy 1; (c) each flux lasted for 24 hours in Strategy 2; (d) each flux lasted for 3 – 4 days in Strategy 3

When the flux was increased to 10 LMH, the TMP increased rapidly again from 1.3 to 2.1 KPa; this TMP oscillation could have been caused by an “unstable” fouling layer which was scoured off the membrane with gas and then reformed at very low TMPs. When the flux was increased to 15 LMH, the TMP stabilized at around 2.1 KPa, which is still a very low pressure drop. With further flux increases, although there was some increase in TMP initially when the flux was changed, it was stable over time at around 2.1, 2.4 and 2.5 KPa for fluxes of 20, 22, and 24 LMH, respectively. However, the TMP profile changed at 26 LMH where a TMP gradient (dTMP/dt) of 0.0105 KPa/min was observed over the 20 mins, higher than at any previous flux. Similarly, at fluxes of

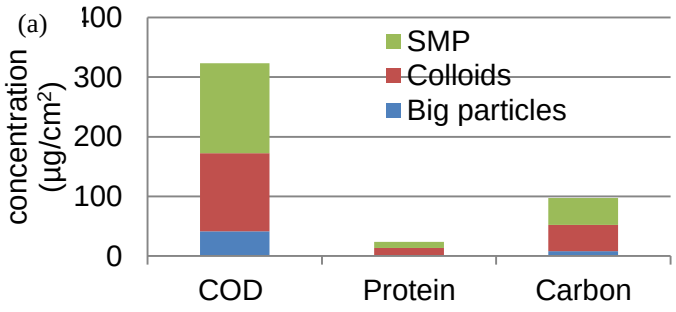
28 and 30 LMH the TMP increased steadily with a similar dTMP/dt slope of 0.0105 KPa/min. When the flux was increased to 33 and 36 LMH, a sharp increase in the TMP gradient occurred indicating a rapid fouling stage. Reversing these steps every 20 mins relaxed the membrane fouling, and over time the TMP decreased to 1.9 KPa. Interestingly, this TMP was identical to the initial TMP and hence there was no irreversible fouling. Due to the unstable TMPs in the initial stage, this experiment was repeated with the fouled but relaxed membrane from the first run (Fig 1b). The TMP profile was similar except in the initial stages at 5 and 10 LMH, where the TMP stabilized around 1.9 and 2 KPa, respectively. Therefore, a relaxed membrane was more stable than a chemically clean membrane when operated under low fluxes. The TMP at 24 LMH plateaued, while a slight slope was found at 26 LMH (dTMP/dt=0.083 KPa/min), which may not be statistically significant.

Longer term operation of the SAMBR to assess critical flux revealed more details about the TMP change under defined fluxes (Fig 1c). At 20 and 24 LMH the TMP fluctuated substantially but was relatively constant over time, but at 26 LMH the TMP increased steadily over time (dTMP/dt=0.02 KPa/h). Therefore, based on steps of 2 LMH, 26 LMH was the flux one step above the “critical flux”, thus the critical flux of the SAMBR was 24 LMH. The last experiment with a constant flux lasting for 3 - 4 days showed that there was an increase of about 0.5 kPa in the TMP for every increment in the flux before a flux of 20 LMH. However, at a flux of 25 LMH, the TMP increase rate became higher from the second day, and jumped from 6 to 8.3 LMH on the fourth day. Therefore, 25 LMH was slightly higher than the “critical flux”, and caused severe membrane fouling after 3 days.

3.2. Characterization of the foulants on the membranes

At the end of strategy 2 (Figure 1c), the fouled membrane was taken out for physiochemical analysis. SMPs and colloids in the form of COD accounted for about 46% and 42% of the supernatant prepared from dissolving the membrane foulants, respectively (Fig 2a), while the balance (12%) was larger colloids (>1μm). Size Exclusion Chromatography (SEC) showed that the reactor supernatant and the effluent were rich in SMPs with molecular weights (MW) between 498.6 and 905 kDa, while the foulant layer was rich in SMPs greater than 1,522 kDa (Fig 3). It is probable that once the fouling layer formed on the membrane, the large SMPs were retained and stayed on the membrane, while the small SMPs passed through the membrane. SEC analysis of the retentate and effluent demonstrated that the retentate was rich in SMPs greater 134 KDa, while the effluent contained very few SMPs larger than 1,522 KDa; thus the SMPs larger than 1,522 KDa were retained by the microfiltration membrane (data not show). Therefore, the colloids and the high MW SMPs contributed most to membrane fouling.

TOC analysis of the foulants for strategy 3 indicated that when the flux increased above the critical flux, the fouling SMPs increased significantly, but the colloids and other organics only accounted for a small portion of the foulants. Colloids were one of the main foulants in Strategy 2 when each constant flux run was for 24 hours, while the colloid concentration was much lower in Strategy 3 for each run of 3 – 4 days. This might because under longer operation times, the colloids on the membrane might have been biodegraded when there was a cake layer containing many microorganisms formed on the membrane. So from the long term perspective, SMPs were much more important in causing the membrane fouling.



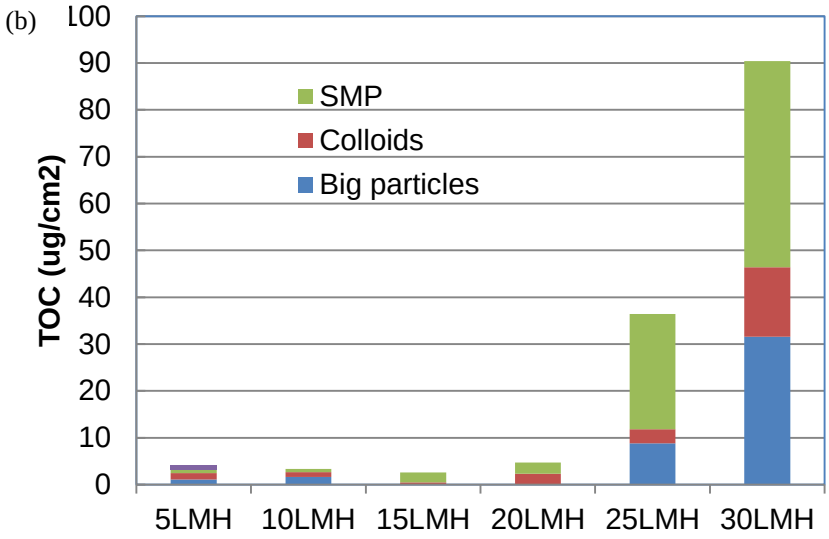


Figure 2: The organic concentration of the supernatant prepared from the membrane foulants (a) at the end of strategy 2 (b) at the end of every flux in strategy 3

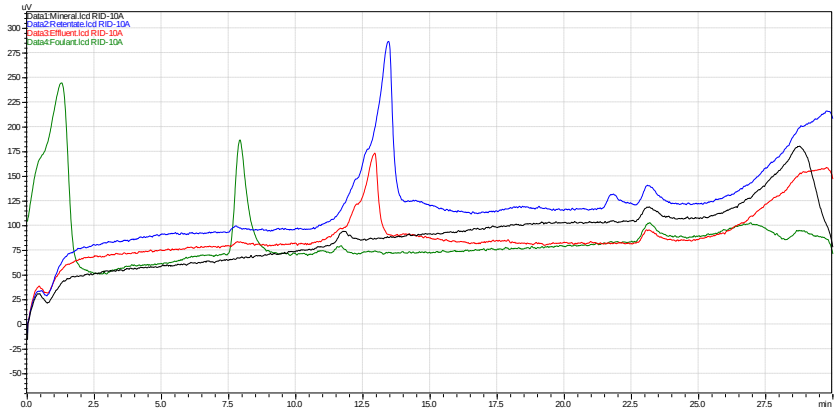


Figure 3: SEC chromatography of the retentate (blue), effluent (red) and foulants (green)

4. Conclusions

(1) The "critical flux" of the SAMBR was found to be 24 LMH despite using 3 very different fouling strategies, although all other parameters were kept constant.

(2) SMPs accounted for about 46% of the overall organics of the supernatant prepared from solubilising the membrane foulants, and SMPs with MWs greater than 1,522 KDa were retained on the membrane, which contributed most to the membrane fouling.

(3) The supernatant prepared from dissolving the membrane foulants were rich in colloids during short term operation, but this reduced during longer term operation.

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