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## **Development and testing of a fully gravitational submerged anaerobic membrane bioreactor for wastewater treatment**

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### **Abstract**

A gravity-operated submerged anaerobic membrane bioreactor (SAnMBR) was set up in order to test its principle of operation as an alternative to conventional pumped permeation of the membrane. This operating mode allowed the membrane flux rate to be measured accurately whilst maintaining a constant transmembrane pressure (TMP), and allowed small transient variations in flux rate to be observed. The reactor was operated at 36 °C for a period of 115 days using a nutrient-balanced synthetic substrate with a high suspended solids concentration. Membrane cleaning was in-situ by a gas scouring system using recirculation of headspace biogas. With an initial TMP of 7.0 kPa the membrane flux slowly decreased due to membrane fouling and had not reached a constant value by day 71. The results indicated that the system was still acclimatising up to 50 days after start-up; but from that point onwards performance parameters became much more stable. A constant flux of 2.2 L m<sup>-2</sup> hour<sup>-1</sup> was achieved over the last 45 days after the TMP was reduced to 2.3 kPa. The stable flux was maintained over this period and the loading raised to 1g COD L<sup>-1</sup> d<sup>-1</sup> by increasing the influent strength. Under these conditions the average COD removal efficiency was 96% and the specific methane potential (SMP) was 0.31 L CH<sub>4</sub> g<sup>-1</sup> COD removed.

**Keywords** Anaerobic digestion, AnMBR, membrane fouling, sustainable membrane flux, wastewater treatment

## 1 Introduction

Anaerobic membrane bioreactors (AnMBR) were first introduced in the 1980s and provide an effective method of: solid-liquid separation and biomass retention [1, 2]; decoupling hydraulic retention time (HRT) and mean cell residence time (MCRT); achieving a high effluent quality; reducing sludge production; and having the potential for net energy production [2]. The main limitation on their widespread application is membrane fouling, which reduces the flux by decreasing the overall membrane permeability [3]. Membrane flux is therefore considered the main parameter for evaluation of the performance and determination of the economic feasibility of these reactors [4]. Alternative methods for assessment of AnMBR membrane performance may contribute to finding operational regimes that establish the best balance between fouling, membrane flux, cleaning frequency and other operational parameters [5]. Smith *et al.* [6] suggested that future AnMBR research should focus particularly on developments that reduce energy demand, and on the relationship between HRT, MCRT, treatment performance and membrane fouling, which is complex and poorly defined in the literature.

In AnMBR two configurations are employed. In the first, the main membrane is housed in a separate vessel to the main reactor and the digester mixed liquor is circulated through it as a side-stream; in the second, the membrane is submerged in the main reactor itself and the mixed liquor circulates around it. While the external configuration can achieve high fluxes due to the relative ease of maintenance and cleaning, its major reported limitations are: it is energy intensive; mixing and shear caused by the pumping system can lead to a reduction in particle sizes; and the release of soluble organics can lead to high volatile fatty acid concentrations that may inhibit methanogenesis [7]. The advantages of a submerged membrane configuration are that it is less energy intensive, and the biomass in the reactor is subjected to lower shear. The major disadvantage is that the membrane is less accessible for cleaning, and in situ methods such as gas scouring need to be employed [7]. Experience of working with submerged AnMBRs is still limited, however, and there is as yet insufficient evidence to support their economic superiority at a large scale over external membrane systems [7].

In submerged systems the most common method of operation is to draw permeate through the membrane by using a pump to reduce the pressure in the membrane lumen. As the membrane pores block, this pressure must be increased to maintain a constant flow. In this case membrane performance can be evaluated by measuring the increase or decrease in the transmembrane pressure (TMP) under a constant flux rate. It is also possible, however, to operate a MBR by gravity permeation, as has been demonstrated in aerobic systems [8, 9, 10, 11]. This relies on a head differential between the inlet and the outlet to the next downstream process. Using this method it is possible to generate a pressure similar to that applied in pumped systems [12]. Operation of an AnMBR by gravity could reduce the parasitic energy requirement for reactor operation [9, 10], although the pumping component may be small compared to the energy required for membrane cleaning e.g. by bubble scouring. Martin *et al.* [13] estimated the pumping energy requirement can represent up to 5% of the total energy consumption in pumped SAnMBRs.

Operating the reactor in this way with a fixed head differential gives a constant TMP, but the flux may be variable, as this changes in response to changes in membrane permeability due to fouling [14,p.32-33]. It is possible, however, that a steady state condition could be reached at which there is a constant flux at a constant gravitationally-induced TMP. What is not known is whether the flux rates under these conditions would be comparable to those in pumped systems. The concept of operating in gravitational mode at a constant TMP also provides an alternative method for assessing membrane performance as a result of fouling. In this case the rate of change of flux provides a measure of the rate of fouling to the point where a steady state is achieved. Although gravitational operation of submerged AnMBRs (SAnMBR) has been suggested by Hong [15], no work appears to have been carried out to date to test or further develop this concept. The work reported here was a preliminary study intended to assess the concept and to establish the likely steady-state flux rates that might be achieved, using a synthetic substrate that was formulated to have a relatively high concentration of suspended solids. The work used a flat plate membrane with gas scouring to maintain membrane permeability, and thus to provide reproducible comparative results over a range of induced TMP values. The use of a synthetic substrate allows experimental runs to be carried out over long durations without changes in the properties of the influent feed. This eliminates the substrate variability that can make interpretation of results difficult; and is thus considered justifiable at an early stage in this type of research [1, 2], as it gives a greater degree of control where a specific aspect of operation is under investigation. In this case the main aim of the work was to investigate rates of membrane fouling under constant TMP and to establish achievable steady state flux rates in a model system.

## **2 Methods and materials**

### **2.1 *Reactor design and function***

The experimental set-up and reactor design are shown in Figure 1. The SAnMBR was constructed in PVC with a Perspex front panel which allowed observation of the water level, solids deposition, and general functioning. The liquid depth inside the reactor was kept at  $61 \pm 0.5$  cm, giving a working volume of 9.1 litres. A headspace was provided above the liquid level to prevent foam bridging between the liquid and biogas outlet; this had a volume of 2.6 L and a depth of 20 cm. The membrane was a Type 203 cartridge (Kubota Co., Japan) which is manufactured from two flat-sheet chlorinated polyethylene membranes mounted on a support frame to provide a lumen from which the permeate can be withdrawn. The membrane cartridge had an effective surface area of  $0.113 \text{ m}^2$  and a nominal pore size of  $0.4 \text{ }\mu\text{m}$ , giving a membrane packing density of  $0.012 \text{ m}^2 \text{ L}^{-1}$ .

The membrane was mounted between two vertical PVC baffles each 7 mm from the membrane surface. These formed an inner upcomer section and two outer downcomer sections, as shown in Figure 1b. The membrane was cleaned by recirculating biogas through a 6 mm tubular stainless steel sparger with four 2 mm holes spaced evenly along its length. The sparger was mounted 15.5 cm from the base of the membrane, and a continuous bubble curtain was maintained using a 12 V DC diaphragm pump (AIRPO<sup>TM</sup>, UK) to provide a gas flow rate of approximately  $0.6 \text{ L min}^{-1} \text{ L}^{-1}$  reactor or  $48.7 \text{ L min}^{-1} \text{ m}^{-2}$  membrane. The gas flow rate was initially set and periodically verified using a rotameter. Because of the positioning of the baffles,

the biogas sparging system also acted as a gas lift pump and provided a means of circulating the mixed liquor.

Feed entered the reactor through a siphon from a constant head gravity feed tank. This tank was continually supplied with substrate from a refrigerated feed storage tank using a peristaltic pump (Cole-Parmer Master Flex L/S, UK), with any excess returned via an overflow. The gravity feed tank thus maintained a constant level in the reactor, automatically compensating for the volume of effluent passing through the membrane. Permeate left the reactor via a siphon induced by the hydrostatic head and the back-pressure of the headspace: the latter was maintained at approximately 0.3 kPa using a water fermentation gas-lock through which the biogas could leave. Changes in liquid levels in the gas-lock also allowed a quick visual check on any pressure differences in the headspace that might occur due to leakage or blockage.

The reactor temperature was controlled at  $36 \pm 0.5$  °C by circulating heated water from a thermo-circulator (MGW Lauda thermo-star, Germany) through a stainless steel heating coil inside the reactor. Both room temperature and reactor temperature were recorded by temperature probes connected to a U3-LV data acquisition device (Labjack, USA). Heat loss was reduced by attaching 10 cm thick high-performance insulation panels to the front and back of the SAnMBR, which were removable to allow visual monitoring.

## **2.2 *Inoculum and substrate***

The reactor was inoculated with digestate taken from a mesophilic digester treating wastewater biosolids (Millbrook Wastewater Treatment Works, Southampton, UK), as recommended by Akram and Stuckey [16], and diluted to 50% with tap water. During inoculation the reactor headspace was purged with nitrogen. The substrate used (Table 1) was formulated to have a high suspended solids concentration and a balanced nutrient composition in the form of carbohydrate, protein, fat and mineral salts. It was prepared from concentrates and could be diluted to give a Chemical Oxygen Demand (COD) of the desired strength depending on the purpose of the experiment. The concentrated medium was prepared fresh every morning and then diluted in the day feed tank and refrigerated to maintain it in good condition over 24 hours. The main feed tank, constant head feed tank, and all feed lines were cleaned daily to minimise risks of substrate degradation by extraneous growth. The COD of the feed was measured both on preparation and after 24 hours storage in the tank. The average of these values was used in calculation of parameters such as organic loading rate (OLR) and specific gas production.

## **2.3 *Operation and control***

The experiment ran for a total of 115 days and comprised a start-up period and four experimental phases (EP), as shown in Table 2. During this time process control was achieved by maintaining the volumetric organic loading rate (based on COD) within defined limits, and by regulation of the reactor mixed liquor suspended solids (MLSS) by controlled wastage.

The flow through the reactor was measured by weighing the collected permeate, using a laboratory scale (PCE Instruments UK Ltd, BDM 15) with a capacity of 15 kg and a readability of 0.5 g. During start-up and EP-1 this was recorded for 10 hours per day on weekdays, and 2

hours per day at weekends. From EP-2 onwards, weights were recorded continuously (24 hours per day) at one-minute intervals. Flow volumes were calculated assuming a liquid density of 1 kg L<sup>-1</sup>.

#### 2.4 Performance and stability

The performance and stability of the reactors were assessed by monitoring influent and effluent COD concentrations, MLSS, pH, volumetric biogas production and biogas composition. COD was analysed using the closed tube reflux method with titrimetric determination of the end point [17]. pH was measured using a temperature-compensated meter and glass electrode (Jenway 3310, UK) calibrated with buffers at pH 4.0, 7.0 and 9.2 (Fisher Scientific, UK). MLSS concentration was measured according to Standard Method 2540-D [18]. Biogas was collected in gas-impermeable bags: the volume was measured using a weight-type gasometer and reported at a standard temperature and pressure (STP) of 0 °C and 101.3 kPa, in accordance with Walker et al. [19]. Biogas composition was measured using a gas chromatograph (Varian GP-3400, USA) calibrated with a standard gas consisting of 36% CH<sub>4</sub> and 64% CO<sub>2</sub> (v/v) (BOC, UK).

Membrane performance was assessed based on the membrane flux at constant TMP. Membrane flux was calculated using Equation 1

$$J = \frac{Q}{A_M} \quad (1)$$

Where:

$J$  = membrane flux (L hour<sup>-1</sup> m<sup>-2</sup>)

$Q$  = reactor effluent flow (L hour<sup>-1</sup>)

$A_M$  = membrane area (m<sup>-2</sup>)

TMP was calculated from Equation 2 [8], based on the head differential between the inlet and outlet taking into account the back-pressure of the biogas produced.

$$TMP = ((H_{wl} - H_e) + (P_b)) 9.81 \quad (2)$$

Where:

$TMP$  = transmembrane pressure (kPa)

$H_{wl}$  = surface level of the mixed liquor inside the reactor (m)

$H_e$  = level of the effluent outlet (m)

$P_b$  = estimated gas-lock pressure-head (m)

9.81 = conversion factor (1 m pressure head of water  $\approx$  0.1 kgf cm<sup>-2</sup> = 9.80665 kPa)

### 3 Results and discussion

#### 3.1 Operational performance

Average values for the main parameters measured or calculated for each of the experimental phases are summarised in Table 3. Details are shown in Figure 2 and discussed in the sections below.

*Start-up.* The system was operated at a constant TMP of 7.0 kPa. The daily average membrane flux dropped from 20.2 to 11.0 L m<sup>-2</sup> hour<sup>-1</sup> over the first 8 days and then began to stabilise (Figure 2). Mixed liquor was removed at a rate of 100 mL day<sup>-1</sup> resulting in a fall in the reactor MLSS concentration from 21.7 g L<sup>-1</sup> to around 15 g L<sup>-1</sup> (Figure 2).

*EP-1.* In this phase the MLSS concentration was maintained between 14 - 16 g L<sup>-1</sup> and the TMP at 7.0 kPa. The daily average membrane flux gradually reduced from 11.3 to 6.7 L m<sup>-2</sup> hour<sup>-1</sup> over the next 43 days (Figure 2) due to membrane fouling. As a result of this the OLR decreased from 1.9 to 1.1 g COD L<sup>-1</sup> day<sup>-1</sup> and the HRT increased from 7 to 12 hours. COD removal was initially 75% and gradually increased to 96%. The CH<sub>4</sub> content was stable at 80 ± 2% but biogas production was variable, with an initial drop in production followed by a recovery. From day 35, however, there was a gradual fall in production as the load to the reactor decreased due to the falling rate of flux. The specific methane production (SMP) was initially unstable as a result of variable biogas production and COD removal rates, and ranged from 0.19-0.38 L CH<sub>4</sub> g<sup>-1</sup> COD removed. From day 40 SMP stabilised at around 0.27 L CH<sub>4</sub> g<sup>-1</sup> COD removed, indicating the reactor was performing stably in terms of COD conversion.

*EP-2.* On day 53 a deliberate intervention was carried out in an attempt to reduce the flux to a lower and more sustainable value. This involved stopping feeding for 5 hours whilst still drawing permeate from the mixed liquor through the membrane. This gave a sharp but transient increase in the MLSS concentration which accelerated the membrane fouling. The result was observable by a reduction in flux from 6.7 to 5.8 L m<sup>-2</sup> hour<sup>-1</sup> (Figure 2), even though the TMP was maintained at 7.0 kPa. Initially it appeared that a steady state had been established, but flux again began to decline around day 60 reaching a value of 5.0 L m<sup>-2</sup> hour<sup>-1</sup> by day 68. This led to a reduction in OLR from 1.0 to 0.8 g COD L<sup>-1</sup> day<sup>-1</sup> and an increase in HRT from 14 to 16 hours with a further reduction in biogas production, although the SMP remained stable at 0.28 ± 0.02 L CH<sub>4</sub> g<sup>-1</sup> COD removed. To maintain the MLSS concentration at a more or less constant value between day 10 and 68, the amount of biomass removed from the system as a proportion of the total biomass present was equivalent to a calculated mean cell residence time of ~200 days.

*EP-3.* On day 68 a valve was installed in the permeate line in an attempt to provide control over the flux rate by decoupling it from membrane fouling: in effect the valve was meant to reduce the outlet permeate flow relative to the degree of valve closure. This intervention was not successful in giving control of the flux rate, and the reasons for this are discussed in section 3.2.1. The valve was removed on day 71.

*EP-4.* The final intervention to establish a stable membrane flux involved a large reduction in TMP from 7.0 to 2.3 kPa. This resulted in an instant decrease in membrane flux from  $4.2 \text{ L m}^{-2} \text{ hour}^{-1}$  to  $2.2 \pm 0.08 \text{ L m}^{-2} \text{ hour}^{-1}$  (Figure 2), which remained constant for the following 44 days until the end of the trial on day 115. The reduced flux gave a HRT of 37 hours corresponding to a very low OLR of  $0.4 \text{ g COD L}^{-1} \text{ day}^{-1}$ . During this phase no attempt was made to control the MLSS, and no wastage took place. As a consequence of the low OLR and extended HRT the MLSS fell to around  $12 \text{ mg L}^{-1}$ , and biogas production decreased to less than  $0.17 \pm 0.01 \text{ L L}^{-1} \text{ day}^{-1}$ . SMP in the first part of this phase increased to  $0.46 \text{ L CH}_4 \text{ g}^{-1} \text{ COD}$ , probably due to the natural reduction in MLSS concentration in response to near-starvation conditions and the internal release of COD through endogenous decay of the biomass. In hindsight, the severe reduction in TMP was far greater than needed to establish steady state flux conditions at the applied load. Rather than increasing the TMP again, the OLR on the system was raised by increasing the COD of the feed substrate, first to  $0.75 \text{ g COD L}^{-1} \text{ day}^{-1}$  on day 86 and then to  $1.0 \text{ g COD L}^{-1} \text{ day}^{-1}$  on day 100. The MLSS responded to these increases in load and the decline in concentration was reversed, albeit slowly. Biogas production increased stepwise with each increase in OLR and the SMP reduced, gradually returning to its previous value of  $\sim 0.28 \text{ L CH}_4 \text{ g}^{-1} \text{ COD removed}$ . This is below the maximum theoretical value of  $0.35 \text{ m}^3 \text{ CH}_4 \text{ kg}^{-1} \text{ COD}$  [20], indicating that a proportion of the methane was leaving in the liquid phase. As no MLSS wastage took place during this phase the calculated MCRT increased, but would not have reached a steady state value within the time span of the trial. At the end of this phase the reactor was operating very stably with an OLR of  $1.0 \text{ g COD L}^{-1} \text{ day}^{-1}$ , a membrane flux of  $2 \text{ L m}^{-2} \text{ hour}^{-1}$  at a TMP of 2.3 kPa, a COD removal of 97 % and a SMP typical of the range reported for SANMBRs working at around this COD concentration [21, 22, 23].

As shown in Table 3, the methane content of the biogas produced was always between 73-83%. This was in agreement with Lin et al. [2] who noted that biogas produced in AnMBR generally contains between 70-90%  $\text{CH}_4$ . The pH remained relatively constant throughout the experimental phases (Table 3) as the feed was sufficiently buffered due to its relatively high nitrogen content, and there was thus no requirement to add sodium bicarbonate ( $\text{NaHCO}_3$ ) to prevent acidification, either manually or using an automatic doser [21, 24, 25, 26].

### **3.2 Membrane performance and fouling phenomena**

The critical membrane flux is defined as the permeate flux above which irreversible fouling appears. For an MBR operating in pumped permeation mode the TMP is increased as fouling occurs to maintain a constant flux. This flux is set below the critical value and is defined as the sustainable membrane flux for the fouling control mechanism in operation [14]. When operating in a gravitational mode the same principles are applied, but in this case the TMP is controlled by the hydrostatic head applied across the membrane. The same definitions apply for critical and sustainable flux. For AeMBR in continuous operation a critical pressure head has also been defined: this is the minimum head that must be applied if irreversible membrane fouling is to be prevented [27]. In the experimental system a sustainable membrane flux of  $2.2 \text{ L m}^{-2} \text{ hour}^{-1}$  was maintained over a 44-day period (Figure 3a) with a hydrostatic pressure head of around 2.3 kPa. Further work is needed, however, to determine the influence of other factors

on this parameter in addition to those associated with the membrane cleaning system. The study reported here is the first on the operation of a SAnMBR and thus the TMP values applied were selected without prior operating knowledge; likewise it was not known what sustainable flux could be achieved for this design of reactor and its gas scour membrane cleaning system. The operational changes between the four experimental phases were attempts to reach a sustainable flux condition within a limited experimental duration. If the original TMP of 7 kPa had been maintained it is likely that a sustainable flux would have been achieved, but its value cannot be calculated from the data available. Ideally a series of different TMPs would be tested until steady state 'sustainable flux' conditions were achieved, and an empirical relationship established: this however would still be dependent on other operating conditions. The effect of these on the system and on flux rates has not been extensively researched: in this study the impact of OLR was investigated in *EP-4*. The results (Figure 2) indicate that a constant flux could be maintained irrespective of the organic load applied, but the sustainability of this constant flux was not fully tested. The MLSS concentration increased as a result of the increased OLR, but only minimally, indicating that the system had sufficient metabolic capacity to be able to degrade the additional COD load. As the system was operated without biomass wastage the food-to-mass ratio remained constant, and further increases in COD load would be expected to lead to a proportional increase in MLSS. The system is further complicated by the effect of endogenous respiration, with long operating periods potentially required for the establishment of stable conditions due to the very high MCRT (calculated as 600 days for *EP-4*). The MCRT controls the sludge growth rate and in many biological systems this is known to influence the production of extracellular polymeric (ECP) materials, which in turn may affect membrane fouling [7]. The current work did not investigate this or other microbiologically mediated factors that may contribute towards membrane fouling, but the importance of these and of the parameters controlling them should not be overlooked [7, 22, 28, 29].

Within the literature there is a wide variety of reported data for SAnMBR, all of which have used pumping to maintain a constant flux, so direct comparison with gravity permeation systems is not possible. The long-term constant flux of  $2.2 \text{ L m}^{-2} \text{ hour}^{-1}$  is, however, comparable to a constant flux of  $1.25 \text{ L m}^{-2} \text{ hour}^{-1}$  reported by Hu and Stuckey [21] at a TMP of 2.5 kPa. For higher operational fluxes of 10 and  $15 \text{ L m}^{-2} \text{ hour}^{-1}$ , they obtained TMPs of 29 and 40 kPa, respectively; many times higher than the maximum TMP of 7.0 kPa employed in this study. Furthermore, the MLSS in their study was three times lower ( $4.1 \pm 0.3 \text{ g L}^{-1}$ ) than the range for sustainable flux during this study ( $12.3 - 14.2 \text{ g L}^{-1}$ ). Similarly, Huang *et al.* [22] reported TMP up to 30 kPa when operating SAnMBR at different MCRT and membrane fluxes between 5 and  $8 \text{ L m}^{-2} \text{ hour}^{-1}$ . Further comparable results for flat sheet SAnMBR can be observed in the review carried out by Skouteris *et al.* [30].

In principle the flux induced by a gravitational system should be the same as that for a pumped system when the TMP values are the same and all other conditions are equal. The limitation in a gravitational system is the maximum TMP value that can be induced, and this is dependent on the hydrostatic pressure head which is determined by the engineering design. For example some full-scale Kubota AeMBR use vertically-stacked membrane cassettes to create additional



hydrostatic head; this of course is only energetically more favourable where additional head is available in the system upstream of the reactor. While absolute values of flux are dependent on the membrane cleaning system, the nature of the wastewater substrate, and the characteristics of the biomass, the results suggest that a gravitational system could be an alternative to pumped systems if additional head is available and measures are developed to incorporate this into the design, as they have been for AeMBRs.

Until steady state conditions are established in the system, the influent flow and load will decline as these are regulated by the achievable membrane flux. Once a constant flux is achieved, however, the system becomes self-regulating, avoiding the need to couple permeate pump flow to influent flow: a gravitational system could thus be simpler to operate and control than a pumped system.

### 3.2.1 *Factors influencing membrane flux*

A continuous detailed log of membrane flux was achieved by recording effluent weight every minute. This showed (Figure 3a) a variation in flux throughout the day with observable oscillations that could be as a result of small environmental changes. The effect of temperature is shown in Figure 3b, where it can be seen that there is a small increase in reactor temperature at the beginning of each day as a result of replenishing the feedstock; with the associated lag in cooling, and also an increase in laboratory ambient temperature during the day. The 25-point moving average for flux mirrors this change in temperature. Temperature is known to affect the viscosity of the substrate, and may also affect the headspace partial pressure and therefore the hydrostatic pressure head on the membrane. Previous research on gravity permeation AeMBRs has shown that temperature has a significant effect on the flux in long-term operation [27].

The effectiveness of biogas scouring in membrane cleaning can be seen in Figure 3c when the biogas recirculation diaphragm pump failed on day 78. This resulted in an immediate and severe flux reduction. Once the pump was replaced there was a rapid increase in membrane flux to a value which was initially higher than that prior to failure, which rapidly returned to the constant flux previously achieved. The membrane fouling as a result of this failure was therefore not deeply embedded, and did not result in critical flux conditions. The general condition of the membrane was assessed visually at the end of the experiment. Figure 4a shows the membrane before and after rinsing with running water: it can be seen that severe fouling occurred only at the centre of the sheet, and was the same on both sides of the membrane cartridge. It is clear that the gas scouring could be improved by better bubble distribution, and this in turn would change the constant flux rate achievable.

Restricting the flow of effluent from the lumen by inserting an in-line valve failed to provide control over the membrane flux as desired, but it did show an interesting result (Figure 5a). It was anticipated that closing the valve would give a lower and stable permeate flow. What was observed was a continuous fall in permeate flow, the rate of which depended on the degree of valve closure, and which did not stabilise over periods of a day or more. When the valve was opened the permeate flow returned to its previous value, indicating that the restriction had not

increased the degree of membrane fouling. The loss of flux was attributed to the accumulation of dissolved biogas inside the membrane, as whenever the valve was re-opened a considerable amount of biogas was released through the permeate line. Throughout the trial occasional gas bubbles could be seen in the membrane permeate line (Figure 4b). During start-up and EP-1 the membrane flux was high enough to carry these bubbles out of the lumen with the effluent stream. As the flux decreased there was an increasing tendency for biogas to accumulate inside the cartridge, interfering with the siphon effect of the permeate line and drastically reducing the flow over short periods, until the volume accumulated was sufficient to eject it from the system; at which point the flux returned to the previous value and biogas accumulation started again. This effect can be seen in Figure 5b as a series of peaks and troughs in the membrane flux. The solution to the problem was to reduce the effluent permeate line diameter from 5 mm to 3 mm: this increased the velocity of flow which effectively dragged the bubbles out with the permeate. The problem of gas liquid phase separation in the lumen highlights one of the issues associated with methane oversaturation in AnMBR due to the TMP forcing more methane into solution, and also to localised biogas generation on and within the membrane itself, as discussed by Smith *et al.* [6]. Dissolved methane can represent a significant loss in methane production and energy potential, as well as a source of fugitive greenhouse emissions [31, 32]. This could also be a problem for the operation of a gravitational SANMBR, but on the positive side this reactor configuration could be engineered to recover dissolved methane as a proportion returns to the gaseous state before leaving the system. More research is therefore required to improve the understanding of the gas bubble phenomenon and exploit any advantages it might offer. Dissolved gases were not measured in this study, but the slightly acidic pH of the effluent (Table 3) and the low percentage concentration in the biogas suggests a high proportion of the CO<sub>2</sub> was dissolved.

### 3.4 Conclusions

Although not tested to its full extent, the principle of using a gravitational SANMBR was established and a constant flux of 2.2 L m<sup>-2</sup> hour<sup>-1</sup> was achieved and was maintained over a period of 44 days at a hydrostatic pressure head of 2.3 kPa. The experimental procedure adopted to establish the system could be used to estimate the rate of long-term fouling, and this technique could have future applications in evaluating fouling rates under different head conditions. The experimental system was also sensitive enough to show small transient changes in membrane flux and could be a valuable tool to study phenomena such as temperature change or biomass characteristics on membrane fouling. In practical operational terms the gravitational system may be simpler to operate than pumped permeation, as once a sustainable flux is achieved the inlet and outlet flows are self-compensating. The major disadvantage was the dissolution of biogas in the membrane lumen, but this could also potentially be turned to advantage if the effluent-entrained biogas could be captured rather than escaping: this is more easily facilitated if it has come out of solution rather than remaining dissolved.

The reactor acclimated well to the substrate used, although the initial choice of COD was, with hindsight, too low for the constant flux finally achieved. The results showed that a much greater loading could be applied whilst maintaining operational performance, as indicated by specific methane yield, COD removal, and volumetric biogas production.

To bring gravitational SAnMBR to a practical reality a greater understanding is required of the operational factors likely to change the characteristics of the MLSS, especially where these impact on membrane fouling. Of particular interest is the relationship between sludge growth rate and ECP production. The gravitational system will be useful in studying this as it can accurately quantify very small changes in membrane flux whilst operating at a constant TMP.

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## Tables

Table 1. SWW composition

| Component                        | Quantity | Unit               | Preparation   |
|----------------------------------|----------|--------------------|---|
| Yeast (block bakers form)        | 23       | g L <sup>-1</sup>  | dissolved in 0.23 l of tap water and autoclaved for 15 min. |
| Urea                             | 2.14     | g L <sup>-1</sup>  | added directly  |
| Full cream milk (UHT sterilised) | 144      | mL L <sup>-1</sup> | Added directly  |
| Sugar (granulated white)         | 11.5     | g L <sup>-1</sup>  | Added directly  |
| Blood (freeze dried)             | 5.75     | g L <sup>-1</sup>  | homogenised with 0.2 l of water                             |
| Ammonia phosphate                | 3.4      | g L <sup>-1</sup>  | added directly  |
| Tap water                        | -        | -                  | to make up to 1 litre*                                      |

\*The above quantities are to produce a concentrate with a COD of approximately 50 g L<sup>-1</sup>, which can then be adjusted as required by dilution with tap water.

Table 2. Start-up and experimental phases

| <b>Phase</b> | <b>Duration</b><br>(days) | <b>Objective</b>  | <b>TMP</b><br>(kPa) |
|--------------|---------------------------|---|---------------------|
| Start-up     | 10                        | Start up experiment and stabilise system for the experimental phases                                | 7.0                 |
| EP-1         | 43                        | First insight into fully gravitational SAnMBR operation and understanding of the system functioning | 7.0                 |
| EP-2         | 15                        | Evaluate membrane performance at high TMP   | 7.0                 |
| EP-3         | 3                         | Evaluate performance with a flow restriction at high TMP  | 7.0                 |
| EP-4         | 44                        | Evaluate membrane performance at a low TMP and different OLRs                                       | 2.3                 |



Table 3. Experimental results summary table

| Parameter   |                   | Start-up  | EP-1                       | EP-2                       | EP-3                       | EP-4<br>OLR-1              | EP-4<br>OLR-2              | EP-4<br>OLR-3              |
|---|-------------------|---|----------------------------|----------------------------|----------------------------|----------------------------|----------------------------|----------------------------|
|   |                   | Membrane Flux (J)*<br>(L m <sup>-2</sup> hour <sup>-1</sup> ) | Ave ± SD<br>Range          | 20.2 → 11.2                | 11.4 → 6.6                 | 5.8 → 5.0                  | 4.0 ± 0.36<br>3.5 – 4.3    | 2.2 ± 0.08<br>2.1 – 2.4    |
| Feed COD<br>(mg L <sup>-1</sup> )                                   | Ave ± SD          |   | 551 ± 55                   | 556 ± 44                   | 583 ± 37                   | 587 ± 24                   | 1111 ± 38                  | 1513 ± 45                  |
| Effluent COD<br>(mg L <sup>-1</sup> )                               | Ave ± SD<br>Range |   | 104 → 25                   | 33 → 27                    | 28 ± 1<br>27 – 29          | 23 ± 5<br>14 – 30          | 48 ± 7<br>39 – 56          | 49 ± 8<br>38 – 58          |
| COD removal<br>(%)  | Ave ± SD<br>Range |   | 75% → 96%                  | 95%<br>94% – 96%           | 95%<br>95%                 | 96%<br>95% – 97%           | 96%<br>95% – 97%           | 97%<br>96% – 98%           |
| OLR<br>(g COD L <sup>-1</sup> reactor day <sup>-1</sup> )           | Ave ± SD<br>Range |   | 1.9 → 1.1                  | 0.9 → 0.8                  | 0.7 ± 0.06<br>0.6 – 0.7    | 0.4 ± 0.01<br>0.4          | 0.7 ± 0.01<br>0.7          | 1.0 ± 0.01<br>1.0          |
| HRT<br>(hours)  | Ave ± SD<br>Range | 4 → 7   | 7 → 12                     | 14 → 16                    | 20 ± 2.0<br>19 – 23        | 37 ± 1.3<br>34 – 39        | 37 ± 0.5<br>36 – 38        | 37 ± 0.5<br>36 – 38        |
| MLSS<br>(g L <sup>-1</sup> )  | Ave ± SD<br>Range | 21.7 → 14.8   | 14.7 ± 0.6<br>13.4 – 15.6  | 14.5 ± 0.1<br>14.4 – 14.7  | 14.0 ± 0.2<br>14.2 – 13.9  | 12.8 → 14.2                | 12.5 ± 0.2<br>12.3 – 12.8  | 12.4 → 14.0                |
| Biogas production<br>(L L <sup>-1</sup> reactor day <sup>-1</sup> ) | Ave ± SD<br>Range |   | 0.66 → 0.33                | 0.35 → 0.26                | 0.24 ± 0.02<br>0.22 – 0.26 | 0.17 ± 0.01<br>0.15 – 0.18 | 0.27 ± 0.01<br>0.25 – 0.29 | 0.33 ± 0.02<br>0.30 – 0.37 |
| CH <sub>4</sub> content in biogas<br>(%)                            | Ave ± SD<br>Range |   | 80 % ± 2<br>77 – 85 %      | 79 % ± 1<br>78 – 80 %      | 79%<br>79%                 | 78 % ± 1<br>77 – 80%       | 76 % ± 1<br>73 – 80%       | 75 ± 1 %<br>74 – 76%       |
| SMP<br>(L CH <sub>4</sub> g <sup>-1</sup> COD rem)                  | Ave ± SD<br>Range |   | 0.27 ± 0.03<br>0.19 – 0.38 | 0.28 ± 0.02<br>0.25 – 0.31 | 0.29 ± 0.04<br>0.24 – 0.33 | 0.46 → 0.32                | 0.29 ± 0.02<br>0.26 – 0.32 | 0.26 ± 0.01<br>0.23 – 0.29 |
| pH  | Ave ± SD<br>Range |   | 6.8 ± 0.06<br>6.7 – 7.0    | 6.8 ± 0.03<br>6.7 – 6.8    | 6.8 ± 0.01<br>6.8          | 6.8 ± 0.02<br>6.8          | 6.8 ± 0.06<br>6.7 – 6.9    | 7.0 ± 0.04<br>6.9 – 7.0    |
| MCRT<br>(days)  | Ave               | 76  | 261                        | 203                        | 365                        | 608                        | 608                        | 608                        |

- initial – final
- min – max
- \* membrane flux daily average
- ± One standard deviation

*Variable trend: ascendant or descendent*  
*Stable range of values*

*Only used to show the spread of the data, as most of the reported parameters are not fully independent (membrane flux, effluent COD, biogas production, OLR, HRT, MLSS, SMP and pH)*

## Figure captions

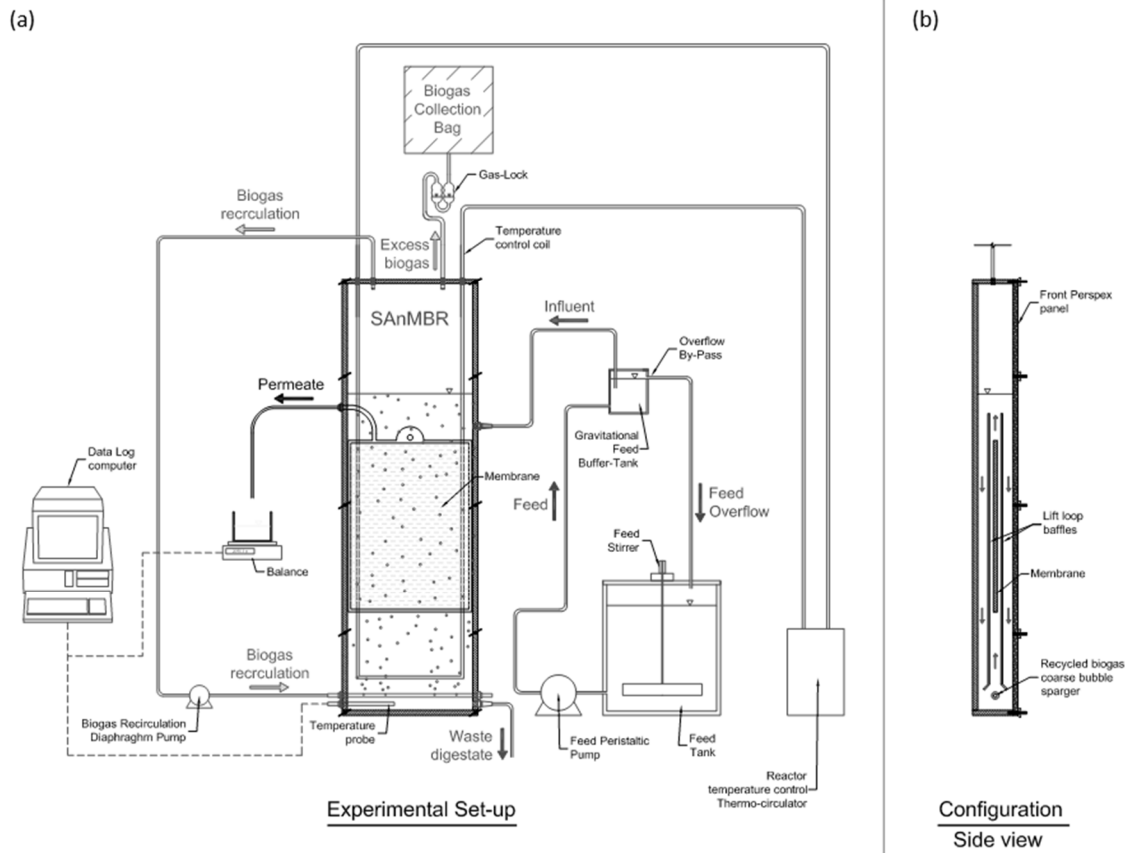


Figure 1. Fully gravitational SAnMBR reactor: (a) experimental set-up schematic; (b) configuration side view

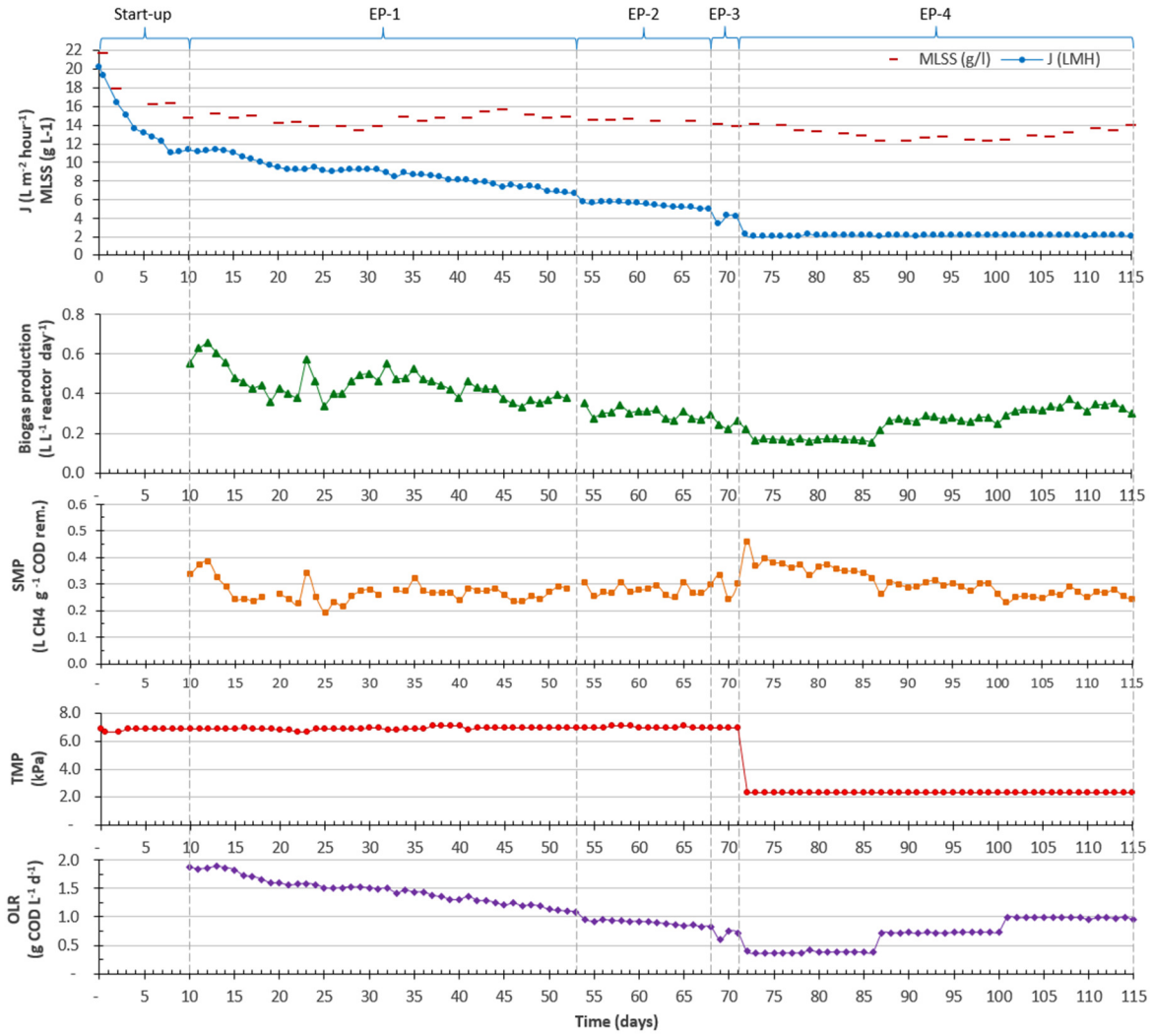


Figure 2. Daily average membrane flux and MLSS concentration, biogas production, SMP, TMP and OLR during experimental period.

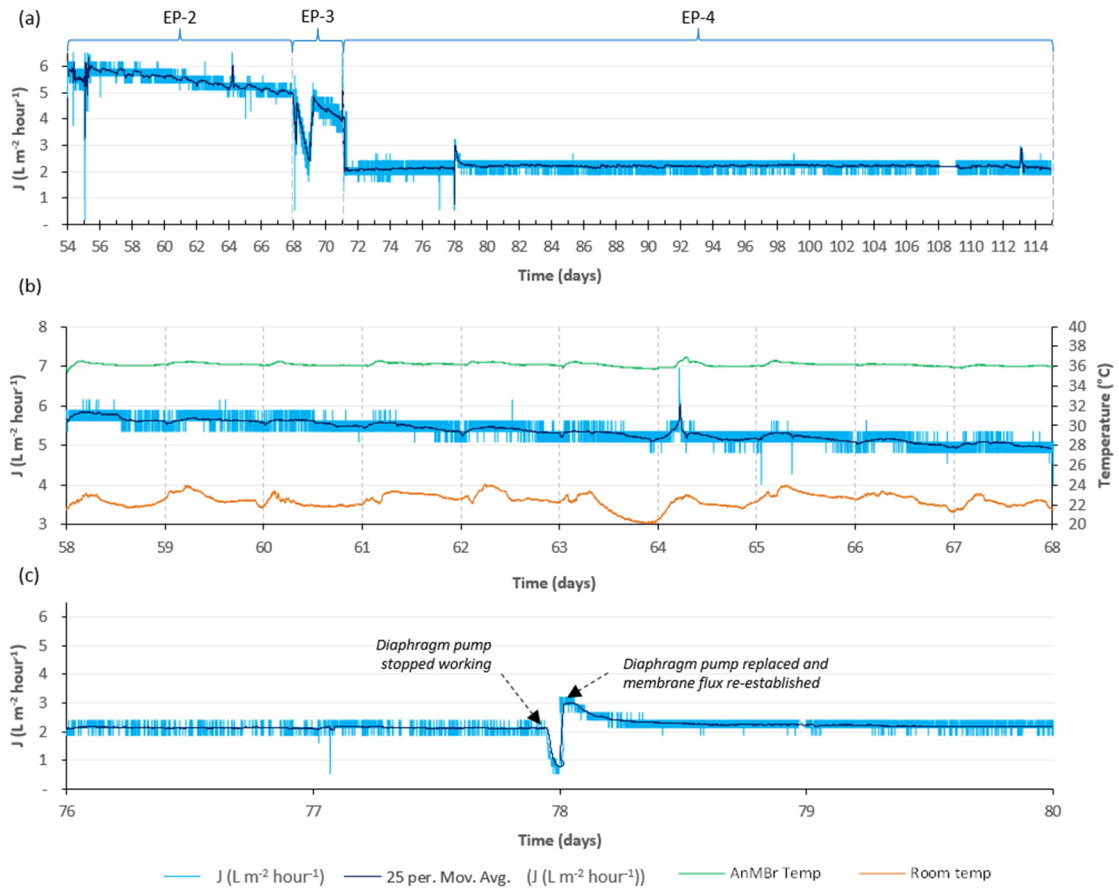
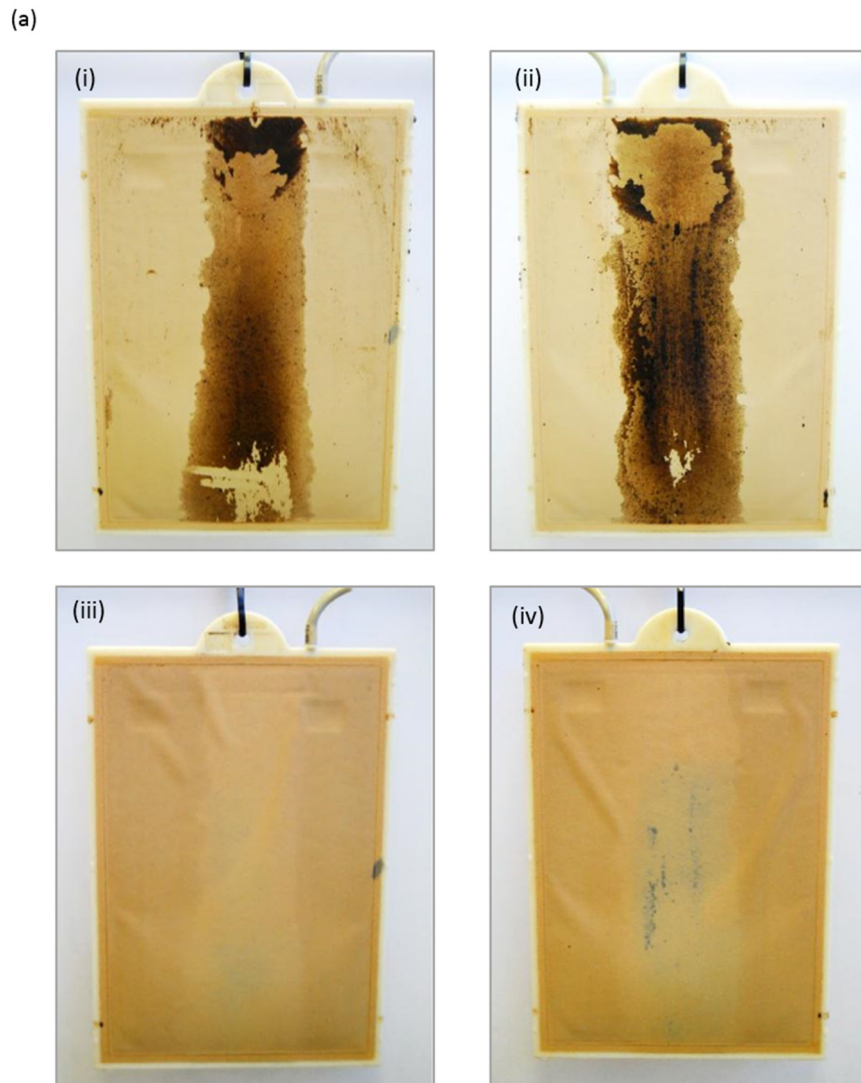


Figure 3. Continuously recorded data for membrane flux: (a) Membrane flux EP-2 – EP-4; (b) Effect of temperature variations on membrane flux, EP-2 (day 58-68); (c) Effect of failure of biogas recirculation pump on membrane flux, EP-4 (days 76-80).



(b)

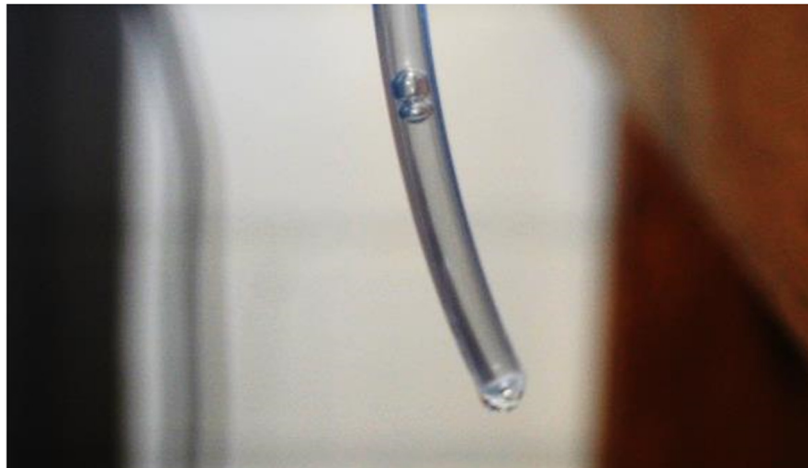


Figure 4. SAnMBR operational details: (a) Membrane at end of experimental run (i) front – before rinse; (ii) back – before rinse; (iii) front – after rinse; (iv) back – after rinse; (b) Biogas bubble in the permeate line.

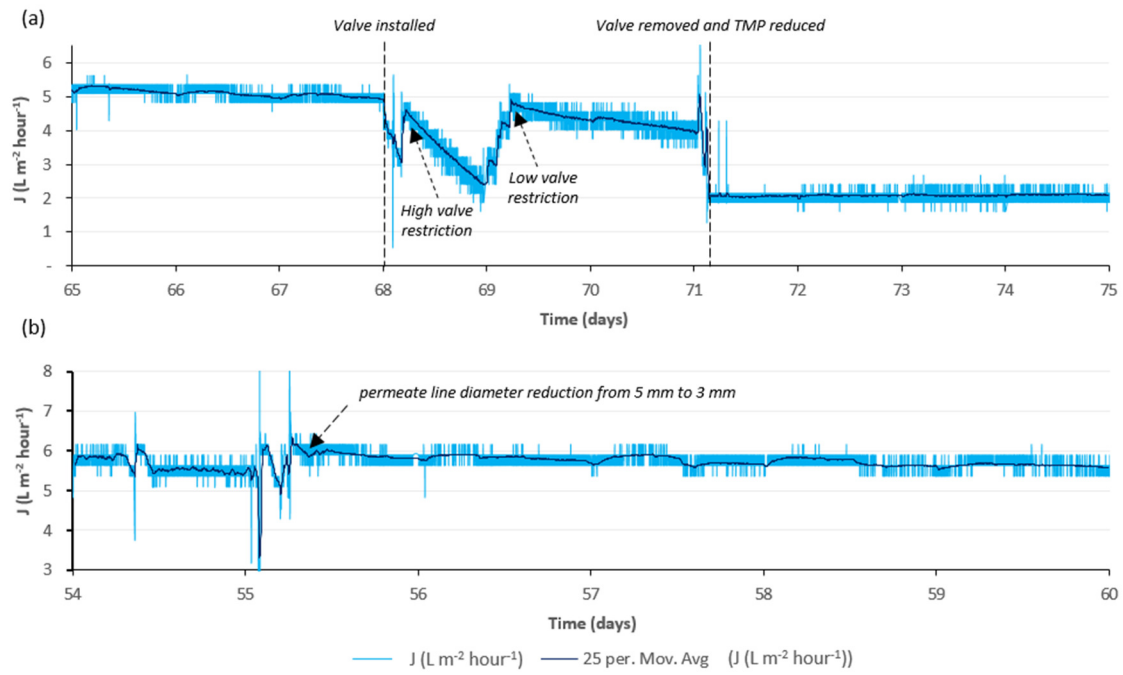


Figure 5. Effect of valve and gas lock on membrane performance: (a) Effect of valve restriction on membrane flux, EP-3 (days 68-71); (b) Effect of biogas accumulation inside the cartridge on membrane flux.