

# Evaluating the Efficiency of Pretreatment to Microfiltration: Using Critical Flux as a Performance Indicator

W. S. Guo<sup>1</sup>, S. Vigneswaran<sup>1\*#</sup>, H. H. Ngo<sup>1</sup>, R. Ben Aim<sup>2</sup>

<sup>1</sup> Faculty of Engineering, University of Technology, Sydney,  
PO Box 123, Broadway, NSW 2007, Australia

<sup>2</sup> LIPE/INSA, Toulouse, France  
<sup>1</sup>Email: wenshan.guo@uts.edu.au  
s.vigneswaran@uts.edu.au  
h.ngo@uts.edu.au

<sup>2</sup>Email: benaim@insa-tlse.fr

## Abstract

In this study, the critical flux was experimentally evaluated for crossflow microfiltration (CFMF) with and without pretreatment. Two different kinds of wastewater (synthetic wastewater with persisting organic compounds and biologically treated wastewater from a wastewater treatment plant) were used. The preflocculation showed a dramatic improvement (nearly 4 times) in the critical flux of the synthetic wastewater (e.g. increased from 100 L/m<sup>2</sup>.h to 380 L/m<sup>2</sup>.h using 0.45 µm microfiltration membrane). The adsorption could increase the critical flux up to 240 L/m<sup>2</sup>.h. When using the biologically treated wastewater, the results indicated that: (i) the pretreatment by adsorption led to 6 times higher critical flux; (ii) the preflocculation alone did not significantly increase the critical flux (there was only a 33% increase with preflocculation) and (iii) the preflocculation combined with PAC adsorption resulted in a very high increase of the critical flux (more than 7 times).

**Keywords** critical flux, crossflow microfiltration, adsorption, preflocculation, pretreatment, powdered activated carbon (PAC)

## 1. Introduction

Application of membrane processes to water and wastewater treatment requires lower investment and operation costs. One of the ways of limiting operation costs is to operate at a constant filtration flux below the critical flux. However, due to the presence of colloids and soluble organics in most raw waters and in secondary treated wastewater, the critical flux is quite low which implies an increase in membrane area and thus in investment cost. The critical flux can be increased by modifying hydrodynamic conditions (e.g. high shear stress), but this results in higher energy consumption and higher operation cost. The influence of additives for modifying the colloidal fraction (flocculants) or entrapping the organic solutes (adsorbents) has to be evaluated, despite the increase in the operating cost.

Membrane systems are becoming increasingly important as cost effective solutions in wastewater treatment and reuse. One of the major drawbacks hindering widespread application of crossflow microfiltration (CFMF) membrane processes in water and wastewater treatment is the gradual reduction in the filtration (permeate) flux below the theoretical capacity of the membranes due to membrane fouling. This membrane fouling is generally caused by the deposition of particles on and within the membrane surface.

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\*Corresponding author: S. Vigneswaran

#Presenting author: S. Vigneswaran

Therefore, an appropriate pretreatment is necessary to improve the performance of the CFMF. The pretreatment can reduce the loading of dissolved organic matter (DOM) on CFMF. Flocculation is one of the pretreatment methods that can improve the permeate flux and remove particles and colloids by CFMF membrane. The flocculation is used to achieve three objectives: eliminating the penetration of colloidal particles into the membrane pores, increasing the critical flux and modifying the characteristics of the deposit [1]. Adsorption is another pretreatment method which can remove dissolved organics, thereby reducing the membrane fouling.

Basically, under the conditions of constant transmembrane pressure (TMP) and cross-flow velocity, the flux in crossflow microfiltration (CFMF) declines to a steady-state value which can be as much as two orders of magnitude lower than the initial or clean water value [2,3]. Howell [4] and Field et al. [5] found significant advantage in CFMF operation when it was operated in sub-critical flux condition. Based on their experimental results, they stated that there exists a critical flux below which a decline of flux with time does not occur; but above this flux, the flux decline is observed. This flux decline can be due to reversible effects such as cake and gel layer, or due to irreversible effects such as adsorption, pore clogging and membrane compactness [6]. The value of the critical flux depends on the hydrodynamics and also on the particle size and their surface and chemical characteristics [7]. This hypothesis of critical flux suggests that when CFMF is operated below a certain filtration flux (critical flux), the fouling of the membrane can be prevented.

In this study, the importance of pretreatment of flocculation and adsorption on flux improvement was evaluated in terms of the critical flux.

## 2. Experimental

In this study, two types of wastewater were used. One is synthetic wastewater with persisting organic compounds and the other is biologically treated sewage effluent from a wastewater treatment plant. The characteristics of biologically treated wastewater used are shown in Table 1. The components of synthetic wastewater are present in Table 2. This wastewater composition was first used by Seo et al. [8] and this wastewater consists of persistent organic pollutants present in the biologically treated sewage effluent. The synthetic wastewater TOC is between 3.8–4.2 mg/L and pH is between 7.6–7.7.

Table 1 Specific characteristics of wastewater used over the experimental period

<b>Wastewater characteristics</b>	<b>Range</b>
Total Organic Carbon (TOC)	1.6–3.8 mg/L
Turbidity	0.8–6 NTU
Orthophosphate ( $\text{PO}_4^{-3}$ )	0.5–12 mg/L
Suspended Solid (SS)	2–15 mg/L

Adsorption and flocculation tests were conducted using a lab-scale batch reactor equipped with mechanical stirrers. Powdered activated carbon (PAC, wood based) was used for adsorption test and ferric chloride ( $\text{FeCl}_3$ ) was used for flocculation test. In both adsorption and flocculation tests, treated wastewaters were settled down before going through CFMF.

Table 2 Constituents of the Synthetic Wastewater

Compounds	Weight (mg/L)
Beef Extract	1.8
Peptone	2.7
Humic acid	4.2
Tannic acid	4.2
(Sodium) lignin sulfonate	2.4
Sodium lauryle sulphate	0.94
Acacia gum powder	4.7
Arabic acid (polysaccharide)	5
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	7.1
KH <sub>2</sub> PO <sub>4</sub>	7.0
NH <sub>4</sub> HCO <sub>3</sub>	19.8
MgSO <sub>4</sub> .3H <sub>2</sub> O	0.71

The schematic diagram of the flat-sheet microfiltration set-up used in the critical flux experiments is shown in Figure 1. The total membrane area was  $3.24 \times 10^{-3} \text{ m}^2$ . The solution was circulated along the surface of the flat-plate membrane in the module and the crossflow velocity was 0.15 m/s. The membranes used are PVDF (modified polyvinylidene difluoride) Minitan-S Microporous Sheets (with pore size of 0.45 and 0.65  $\mu\text{m}$ ). In each experiment, new membrane was used to obtain reproducible results. The biologically treated wastewater was delivered from a stock tank to the CFMF cell. The reject water and filtered water was returned to the feed tank. The initial transmembrane pressure was controlled by two valves and its variation during the filtration was monitored by using a pressure transducer at three points P1, P2 and Pf respectively. During the experiment, the filtration flux at each step was kept constant for at least 40 minutes. The TMP was calculated using the following equation:

$$\text{TMP} = (P1 + P2)/2 - Pf$$

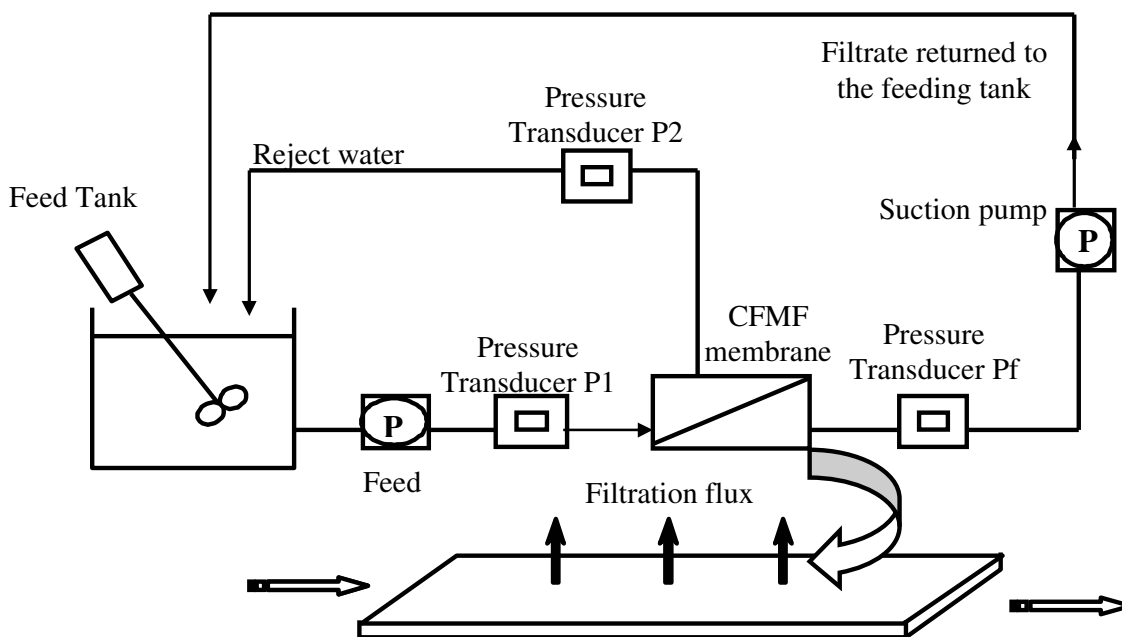
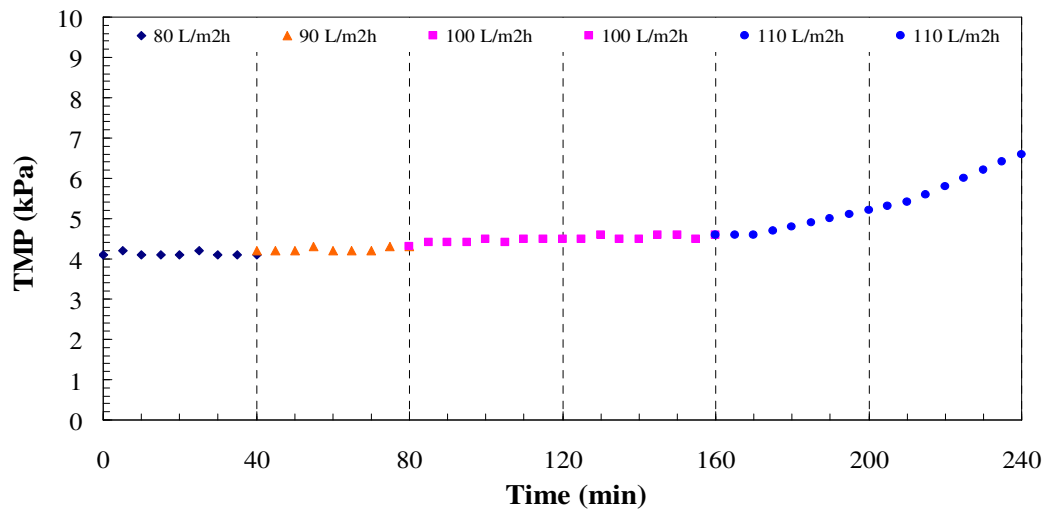


Figure 1 Schematic diagram of the CFMF experimental set-up

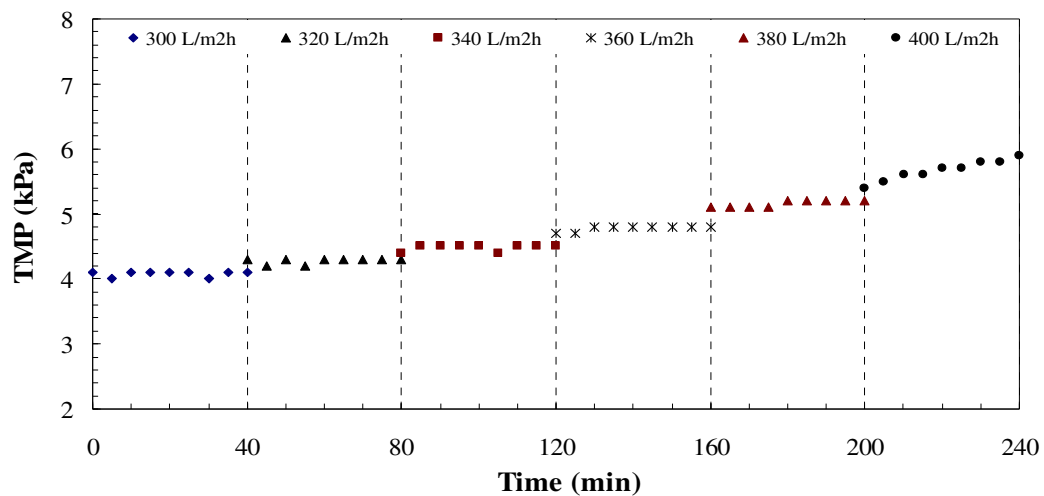
### 3. Results and Discussion

#### 3.1 Critical flux experiments with synthetic wastewater

Figure 2 shows the variation of TMP value for constant filtration flux using the synthetic wastewater without and with preflocculation. Flocculation was operated in batch mode in the following conditions: addition of  $\text{FeCl}_3$  (68 mg/L), agitation during 20 minutes and settling for 20 mins prior to application of the supernatant to CFMF. The critical flux was around  $100 \text{ L/m}^2\text{h}$  for the wastewater without preflocculation (Figure 2a). It increased to  $380 \text{ L/m}^2\text{h}$  when a pretreatment of flocculation was provided (Figure 2b). These results confirm the importance of flocculation in enhancing the critical flux. The flocculation can help in capturing and agglomerating colloids, thereby reducing the membrane fouling by colloids. According to the results, flocculation had more considerable effect on the critical flux than adsorption. Size distribution analyses conducted with the synthetic wastewater with and without flocculation indicated that flocculation can remove all the organics ranging from 30,000–70,000 daltons. It also removed some of the small molecular weight organics (300–2,000).



(a) Without preflocculation



(b) With preflocculation

Figure 2 Constant filtration flux of synthetic wastewater with persisting organic compounds (membrane pore size  $0.45 \mu\text{m}$ ;  $\text{FeCl}_3$  dose = 68 mg/L; crossflow velocity = 0.15 m/s)

The effect of adsorption as pretreatment on critical flux is shown in Figure 3. Here, 2 g/L PAC was added in the wastewater and mixed for 1 hour before the PAC was settled down for 1 hour. With the pretreatment of adsorption, the critical flux increased from 100 L/m<sup>2</sup>.h to 240 L/m<sup>2</sup>.h. With flocculation-adsorption as pretreatment, the critical flux increased more than 5 times (about 520 L/m<sup>2</sup>.h) that of synthetic wastewater without pretreatment. The results indicated that the pretreatment of flocculation-adsorption was able to remove large and small molecular weight organics while enhancing the filtration flux of CFMF and dissolved organic removal. The MW size distribution analysis showed a significant removal of small MW organics between size 300 to 2,000 daltons.

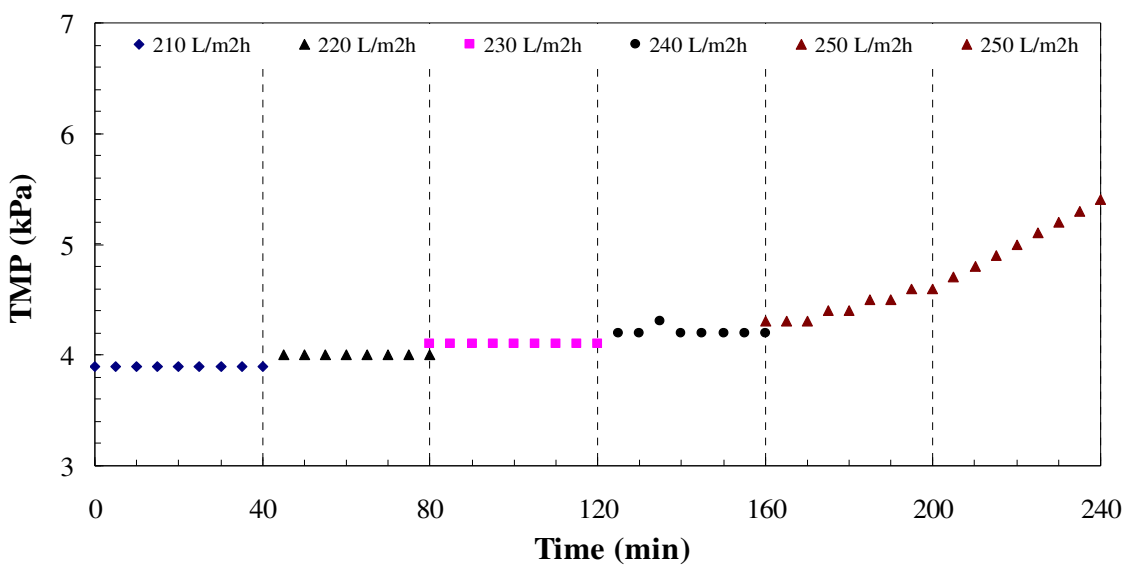


Figure 3 Constant filtration flux of synthetic wastewater after adsorption (membrane pore size= 0.45  $\mu$ m; PAC dose = 2 g/L; crossflow velocity = 0.15 m/s)

### 3.2 Critical flux experiments with biologically treated wastewater

The experiments on critical flux were also conducted with biologically treated wastewater from a wastewater treatment plant. An in-line static flocculator was used as a pretreatment to the CFMF membrane unit (membrane pore size 0.65  $\mu$ m). Adsorption test was carried out using a batch reactor with mechanical stirrers.

As can be seen from Table 3, the critical flux of the biologically treated wastewater was around 150 L/m<sup>2</sup>.h. A pretreatment of flocculation led to an increase in critical flux to 200 L/m<sup>2</sup>.h. However, when wastewater was pre-treated using adsorption, the critical flux increased to 900 L/m<sup>2</sup>.h. Furthermore, when both flocculation and adsorption were used together as pretreatment, the critical flux of wastewater increased dramatically (1100 L/m<sup>2</sup>.h). In the case of biologically treated effluent, adsorption had more significant effect than flocculation. The analysis of MW size distribution with a similar biologically treated sewage effluent indicated that the pretreatments of adsorption and flocculation-adsorption removed the majority of the organics from 300 to 5,000 daltons. The flocculation alone was successful in removing more than 70% of the colloidal effluent organic matter (EFOM) of 3,500 daltons to 0.1  $\mu$ m.

Table 3 The critical flux under different conditions

Experimental condition	Critical flux (L/m <sup>2</sup> h)
Biologically treated wastewater	150
wastewater after flocculation (FeCl <sub>3</sub> : 50 mg/L)	200
wastewater after adsorption (PAC: 2 g/L)	900
wastewater after flocculation (FeCl <sub>3</sub> : 50 mg/L) and adsorption (PAC: 2 g/L)	1100

#### 4. Conclusions

Flocculation as pretreatment to CFMF was successful in improving the critical flux, especially in the case of synthetic wastewater. However, the improvement in critical flux by preflocculation of biologically treated wastewater was less significant (150 L/m<sup>2</sup>.h to 200 L/m<sup>2</sup>.h) comparing to the one with synthetic wastewater. The pretreatment with adsorption led to considerable increase in critical flux of the biologically treated wastewater. With both flocculation and adsorption as pretreatment of the biologically treated wastewater, the critical flux increased more than 7 times. These results confirm that the pretreatment by flocculation and/or adsorption is the key feature to obtain simultaneously higher permeate flux and quasi steady conditions of operation.

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