Critical Flux Determination by Flux-Stepping

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In membrane filtration related scientific literature, often step-by-step determined critical fluxes are reported. Using a dynamic microfiltration device, it is shown that critical fluxes determined from two different flux-stepping methods are dependent upon operational parameters such as step length, step height, and flux start level. Filtrating 8 kg/m^3 yeast cell suspensions by a vibrating 0.45 $\times$ 10^{-6} m pore size microfiltration hollow fiber module, critical fluxes from 5.6 $\times$ 10^{-6} to 1.2 $\times$ 10^{-5} m/s have been measured using various step lengths from 300 to 1200 seconds. Thus, such values are more or less useless in itself as critical flux predictors, and constant flux verification experiments have to be conducted to check if the determined critical fluxes can predict sustainable flux regimes. However, it is shown that using the step-by-step predicted critical fluxes as start guesses, in our case, in constant flux verification experiments for 5 and 1/2 hours, a sustainable flux was identifiable. © 2009 American Institute of Chemical Engineers *AIChE J*, 56: 1739–1747, 2010

Keywords: critical flux, dynamic microfiltration, yeast cells, step length, flux-stepping, bioseparation, membrane separations

Introduction

The concept of critical flux in membrane filtration processes has in the last 15 years gained more and more interest as a way of controlling fouling to achieve sustainable operation. When talking about critical fluxes, sustainable flux regimes are defined as sub-critical flux regions that are sustainable from a constant flux and TMP point of view. By operating at sub-critical flux, the need for membrane cleaning, necessary due to enhanced hydraulic resistance will be reduced. In that way, for example macromolecular transmission in microfiltration can be improved.[1] In 2006, an in-depth review about critical and sustainable fluxes was published by Bacchin et al.[2] In this review it is underlined that three different forms of the critical flux should be distinguished according to the level of hydraulic resistance: The *strong form*, the *weak form*, and the *irreversible form*. The strong and weak forms of the critical flux are well known and described in the literature. In the irreversible form of the critical flux, however, the hydraulic resistance "just" has to be kept below the sum of a membrane resistance, an eventual adsorption resistance, and a reversible fouling resistance not to be exceeded. When using the search words "critical flux," "membrane," or "?filtration," almost 200 papers appear, published since 1979 (using the DADS database, Technical Knowledge Center, Technical University of Denmark, February 2008).

The majority of all critical flux determinations reported in the literature are conducted using step-by-step methods, in which either the transmembrane pressure (TMP) or the flux is stepwise increased, and the response (either flux or TMP) is monitored. This method is easy to apply, which might explain its popularity, and out of the almost 200 mentioned "critical flux papers", the step-by-step method is applied in more than 100 of these papers to determine critical fluxes. However, a main disadvantage of the step-by-step technique is that it only measures the critical flux of the dominant fouling species.[3–5] Furthermore, when using this method, the determined critical fluxes can vary quite much depending on the flux start level J_0, step length sl, and step height sh although, only very few publications actually focus on this. The step height was reported by Le Clech et al.[6] to be the

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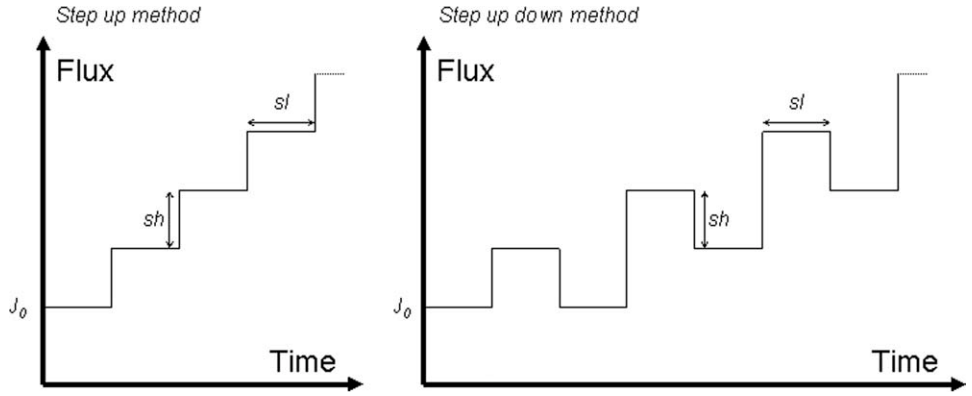

Figure 1. Sketch of two different approaches for critical flux determination.

Step up method and *step up down* method. J_0: Initial flux level, sh: step height, sl: step length.

main parameter impacting the fouling rate (dP/dt), using an MBR system processing sewage, whereas the step length has been reported by Kwon and Vigneswaran[7] not to influence the determined critical fluxes for crossflow microfiltration of latex particles with step lengths of 1200 and 2400 seconds. Neither did Guo et al.[8] detect a difference in the determined critical fluxes in crossflow microfiltration of synthetic wastewater when using step lengths of either 2400 or 7200 seconds. Also, Le Clech et al.[6] only detected minor differences in the determined critical fluxes for step lengths of 300, 600, 900, and 1800 seconds in MBR filtration of synthetic sewage. Only when the step length was increased to 7200 seconds, a significant change in the critical flux could be detected, whereas Choi and Dempsey[9] observed the determined critical fluxes to decrease when the step length was increased from 600 over 1200 to 1800 seconds, using a low pressure membrane system processing raw water.

The aims of this article are (i) to show that one cannot be sure to use a critical flux value based on one single step-by-step measurement to actually determine a critical flux, (ii) to show that a step-by-step determined critical flux has to be verified against constant flux experiments, and (iii) to emphasize that when talking about critical fluxes and sustainable flux regimes, a time period for sustainability has to be assessed. Two different flux-stepping critical flux determination methods are tested using our dynamic microfiltration system. It must be emphasized that we use the term "critical flux" as the outcome of the flux-stepping experiments, although it is later shown that many of these determined values actually are not real critical fluxes able to predict long term behavior. A step up method and a step up down method are tested using different values of the step length to evaluate the response in the determined critical fluxes. Furthermore, the onset of irreversible fouling will be tried to be determined using the step up down method. The experimentally determined critical fluxes are verified against constant flux experiments at, above and below, the average critical fluxes to check if one actually can predict sustainable behavior for an extended period, which in this case is 5 and 1/2 hours. Suspensions of bakers yeast cells are being processed. This medium is chosen to use some sort of a model fluid to simulate fermentation broth or other biological media as we in our earlier work also have added macromolecules to such suspensions.[1] Furthermore, our later results[10] are also obtained using yeast cell suspensions containing different macromolecules. Our system is in many contexts thought to be coupled to treatment of such media.

Flux-stepping concept

When the critical flux is determined by a step-by-step method, the flux is often controlled, and the corresponding TMP is monitored. The flux is increased in steps and at a point, at which the TMP increase exceeds some defined threshold limit, the critical flux is said to be exceeded. However, as pointed out by Bacchin et al.,[2] it would be more correct to talk about this level as a sustainable flux beause the transition detected by this method is often the transition from "slow fouling" to "rapid fouling." Still, we will use the term "critical flux" to emphasize that this is the flux one has to stay below to achieve sustainable filtration. Two different flux-step approaches are sketched in Figure 1.

These two methods are chosen, as they are both well known and often reported in the literature. Important parameters in both methods are the initial flux level (J_0), the step height (sh), and the step length (sl). Le Clech et al.[6] introduced three key TMP related parameters, useful to determine the onset of fouling, for a step up method. In this work, these parameters have also been applied for a step up down method. In Figure 2, the concept is sketched for both methods.

The average of the TMP values for a flux step is denoted P_{avg}, whereas dP/dt is the slope of the linear regression line covering the TMP data of a flux step. The latter parameter is often referred to as the "fouling rate". The sudden increase in TMP in the transition from one flux step to another is denoted ΔP_0 and can be determined as the distance between the linear regression lines for the two particular flux steps at the time of transition. The critical flux is determined from plots of P_{avg}, dP/dt, and ΔP_0 vs. the flux as the point where the curves "break". The advantage of the step up down method is that, in theory, fouling irreversibility can be recognized at a certain flux level as each flux level is "touched" two times. Thus, the onset of irreversible fouling, irreversibility form of critical flux can be identified by plotting the difference in P_{avg}, J, and dP/dt, ($P_{avg,n,ii} - P_{avg,n,i}$), ($dP/dt_{n,ii} - dP/dt_{n,i}$), and ($J_{n,i} - J_{n,ii}$), as function of the flux. This method, distinguishing the reversible fouling and the irreversible fouling, is earlier described by Espinasse et al.[11]

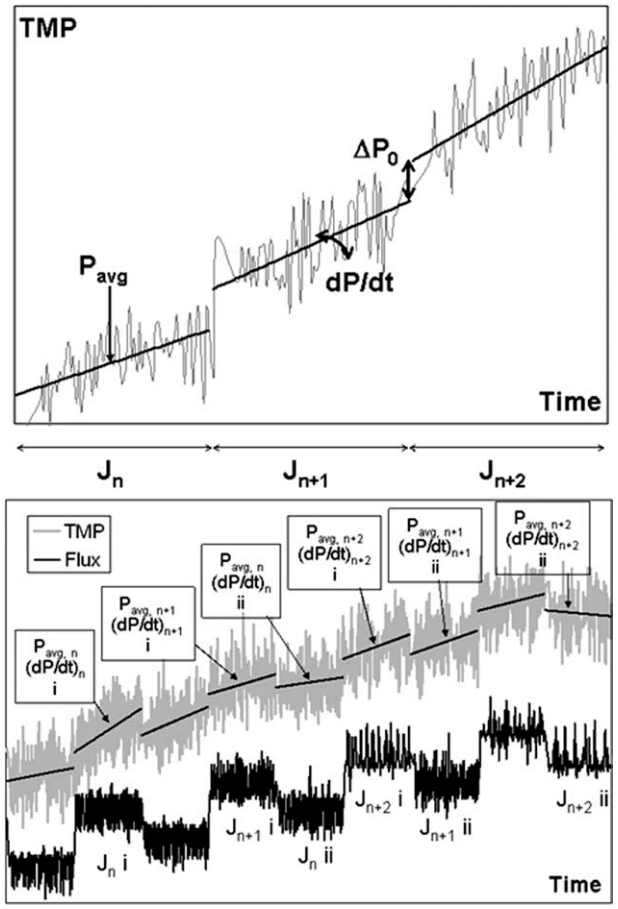

Figure 2. TMP related key parameters.

Upper plot: *Step up* method. P_{avg}, dP/dt, and ΔP_0^6. Lower plot: *Step up down* method. P_{avg} and dP/dt. n: number of the flux step J_n.

Materials and Methods

Apparatus and feed suspensions

The vibrating microfiltration system, often referred to as a dynamic microfiltration system, used in this work consists of a module with 9 hollow fibers placed vertically in a bundle. The system is sketched in Figure 3.

The hollow fibers are all closed in the bottom ends by a steel plate. The top ends of the fibers are, via a permeate gap and the permeate pipe, connected to a suction pump (permeate pump) that sucks permeate through the fibers at a constant rate. Permeate is collected in a beaker on an electronic scale connected to a PC. Permeate is manually poured back to the feed tank when the volume in the permeate beaker is around 100 ml. The permeate pump is controlled by a PC, and the corresponding TMP is monitored and logged by the PC by use of a pressure transducer. The module is placed in a plastic cylinder connected to a feed tank. The feed fluid (3 liters in total) is circulated between the feed tank and module cylinder by a feed pump at very low pumping rate, corresponding to a velocity in the module cylinder below 1×10^{-2} m/s. The temperature is monitored during the experiment by a thermometer in the feed tank, and the temperature is kept constant by a thermostatic regulation on the feed side. The membrane module can be vibrated in the module cylinder at variable frequency and amplitude by a "rotation head," and the frequency and amplitude can be varied independently. Suspensions of bakers yeast cells are filtrated. Solutions contained 8 kg/m^3 dry weight bakers yeast. The suspensions were all buffered to a pH of 6.3 by adding 100 ml of 30 mM phosphate buffer ($KH_2PO_4/Na_2HPO_4{\cdot}7H_2O$), which yield a feed buffer concentration of 1 mM. Relevant membrane parameters are listed in Table 1 together with the module hydrodynamic parameters used in this work.

The skin layer is located on the outside of the fibers, which are made of a polyethersulphone (PES) and polyvinylpyrrolidone (PVP) blend in a 98/2% ratio. PVP is added to make the fibers more hydrophilic as hydrophilic membranes tend to foul less. The average water permeability of the clean membrane module was measured to 2.2×10^{-8} m/(s $\times$ Pa) with a standard deviation of 13% based on 16 measurements. Neither an increasing nor a decreasing tendency of the water permeability was observed during the experimental work. Between each experiment, the membrane module was chemically cleaned with a 2% alkaline solution (P3 Ultrasil-141) for 30 minutes at 50°C.

Improvements of the system have been made compared with the system used and described earlier.[1,12] The number of fibers has been decreased from 54 to 9. The membrane area/module volume ratio has, therefore, been decreased, but the modified configuration makes it much easier to do reparation in case of a single fiber breakage. The liquid level above the module has also been increased from 1×10^{-2} m to 15×10^{-2} m. In the old system, with a liquid level above the module of only 1×10^{-2} m, air bubbles were induced and spread out in the whole module cylinder because of the fast vibrating module. The air bubbles might have disturbed the picture so that the effect of the pure vibrations was not clear. In the new system, with a liquid level of 15×10^{-2} m above the module, no air bubbles are induced at the liquid

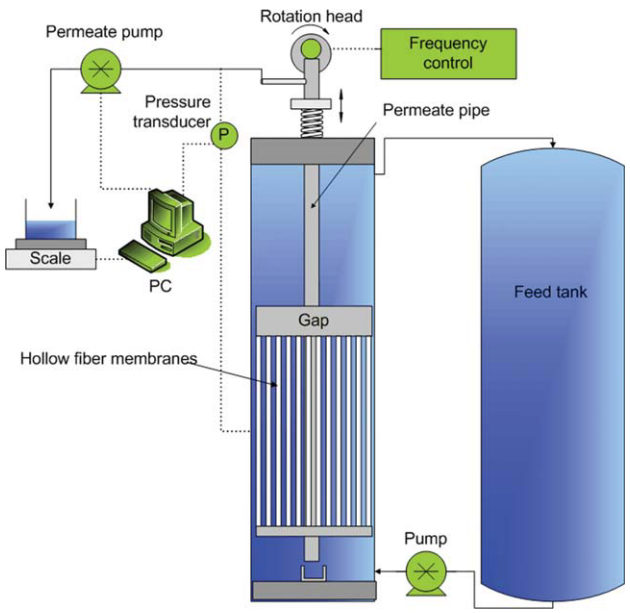

Figure 3. Sketch of the experimental apparatus.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Table 1. Characteristics of the Membrane Module and Hydrodynamic Conditions During the Experimental Runs

Membrane Material	Number of Fibers	Length of Fibers (m)	Total Membrane Area (m^2)	Pore Size (m)	Permeability of Module [m/(s × Pa)]
PES/PVP (98/2%)	9	0.125	0.084	$0.36 \times 10^{-6} - 0.50 \times 10^{-6}$	$2.2 \times 10^{-8} \pm 13\%$
Feed flow velocity (m/s)	**Vibration frequency (s^{-1})**	**Vibration amplitude (m)**	**Average surface shear rate (s^{-1})**	**Temperature (°C)**	**Bakers yeast concentration (kg/m^3)**
9×10^{-3}	20	1.375×10^{-3}	1235	29	8

surface. Though air bubbles are often reported to reduce fouling in MBR systems, we focus on investigating effects of pure vibrations with our system so the elimination of air bubbles is an advantage from that point of view.

Experimental Procedures

All experiments were conducted at the same hydrodynamic and physical conditions as listed in Table 1. The vibration frequency and amplitude of 20 s^{-1} and 1.375×10^{-3} m, respectively, can be calculated into an average surface shear rate of 1235 s^{-1} according to the fluid dynamical equation of our previous work.[12] The mass flow of permeate and TMP were logged every 4th second in all experiments, and the feed recirculation velocity was kept very low, below 9×10^{-3} m/s in the module cylinder between the module and the feed tank. The operational parameters for the critical flux determination experiments are listed in Table 2. Each experimental run was repeated three times. As a limit for acceptable fouling rate (dP/dt), we use a value of 1.1 Pa/s (40 mbar/h), which we have defined and used in a previous study, in which critical fluxes were determined using a step up down method, for the same type of feed suspension.[12] This limit indicates the shift from slow fouling to rapid fouling.

The values of the flux start level, step length, and step height were chosen at the same level as for many earlier reported critical flux studies. Constant suction experiments for 5 and 1/2 hours were also conducted to verify the experimentally determined critical fluxes. Each constant suction experiment was conducted three times. Experiments were conducted at, above and below, the average experimentally determined critical flux:

- Sub-critical flux: $\sim 2.8 \times 10^{-6}$ m/s below the average determined critical flux (Cho and Fane[13] state that extended operation at a fixed "sustainable" flux should be possible as long as the flux is substantially below the nominal critical flux of the dominant foulant).
- At the average determined critical flux.
- Supra-critical flux: $\sim 2.8 \times 10^{-6}$ m/s above the average determined critical flux.

Each experiment was initiated by slowly step-wise increasing the flux until the fixed flux level was reached. Step length of 120 seconds and step heights of 5.6×10^{-7} m/s were used to reach the fixed flux level not to over-foul the membrane by imposing the given flux level immediately. The importance of a slow start-up procedure is underlined by Le Clech et al.[6] who proposed a small initial step height. Also, Chen et al.,[14] working with microfiltration and ultrafiltration of silica particles, stated that slow incrementation of the flux to a given value can result in a significantly lower TMP than the direct application of that flux.

Results and Discussion

Critical flux determination

The critical fluxes were determined in different ways using a step up and a step up down method. The TMP curves were analyzed as sketched in Figure 2. At a certain flux level, for both methods, the increase of pumping rate was not able to increase the flux further, and the limiting flux was reached (around 1.25×10^{-5} m/s). After that, the flux actually decreases when the pumping rate is increased. This is probably because after the critical flux has been reached, cavities are induced in the permeate pump because of deposition, fouling, and probably pore blocking of the membrane which lead to a large increase in hydraulic resistance. The pump is, therefore, not able to maintain the flux. Bacchin[15] has proposed a theoretical link, stating that the critical flux equals 2/3 of the limiting flux, which then will give a critical flux of around 8.3×10^{-6} m/s.

In Figure 4, it is seen how the critical flux is determined based on the dP/dt parameter, fouling rate, when plotted vs. the flux for both methods for a random choice of operational parameters. The uncertainty in the fouling rates is seen as the standard deviation, especially above the critical flux, is very high whereas below this level the fouling rate is relatively constant. The convective forces dragging the main foulant species toward the membrane must exceed the drag forces away from the membrane surface around the critical flux, making the fouling rate curve "break". However, the fouling rate is not zero below the critical flux, which indicates a slow, progressive fouling build-up below the critical flux. Therefore, in our case the critical flux indicates a change in the type of fouling and not the onset of fouling.

Using the method of Le Clech et al.,[6] the critical flux was further determined from plots of the flux vs. the average pressure P_{avg} (TMP), which is shown in Figure 5 for a random choice of operational parameters. The deviation from linearity can be identified as a critical flux. It is, furthermore, seen that the critical flux is of the weak form, according to the definition of the work of Field et al.,[16] as the slope is smaller than the slope of the water flux curve. This, probably, indicates adsorption that changes the permeability of

Table 2. Operational Parameters for the Critical Flux Determination Experiments

	Step Up Method	*Step Up Down* Method
J_0, flux start level (m/s)	1.9×10^{-6}; 4.4×10^{-6}	1.9×10^{-6}
sl, step length (s)	300; 600; 1200	300; 600
sh, step height (m/s)	5.8×10^{-7}; 1.3×10^{-6}	5.8×10^{-7}

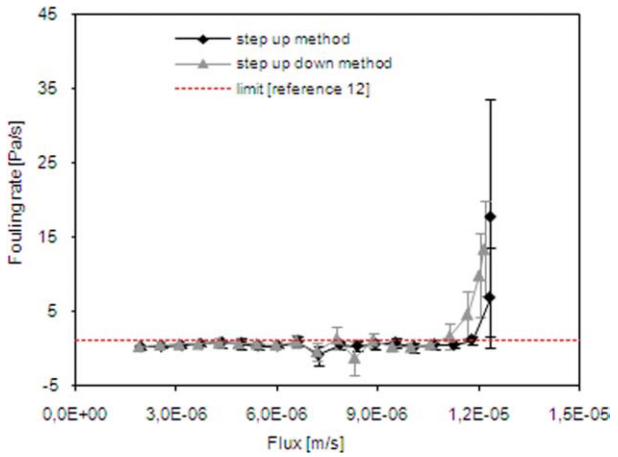

Figure 4. Fouling rate (*dP*/*dt*) vs. flux for one of the *step up* experiment (step length: 1200 s, step height: 5.8 × 10^{-7} m/s, flux start level J_0: 1.9 × 10^{-6} m/s) and for one of the *step up down* experiments (step length: 600 s, step height: 5.8 × 10^{-7} m/s, flux start level: 1.9 × 10^{-6} m/s).

Standard deviation for the three identical experiments is shown with error bars. The fouling rate threshold limit of 1.1 Pa/s is adapted from our previous work[12].

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the membrane. We have earlier determined such irreversible adsorption resistances caused by macromolecular adsorption on ultrafiltration membranes.[17] The decreasing behavior of the flux is probably caused by cavitations in the pump at the largest flux levels.

The critical flux based on the parameter ΔP_0 (Figure 6) is identified at the point where the value (or the standard deviation) undergo a dramatic change. In Figure 6, three step lengths for the step up method are shown. It is seen that using this parameter for identification of the critical flux, with our experimental system, is very uncertain. For a step length of 1200 s, a relatively large deviation in the curve is observed slightly above 1.1 × 10^{-5} m/s, but the deviation could also be assessed just slightly above 5.6 × 10^{-6} m/s if the fluctuation in the curve at that point is defined as being too large. For the step length of 600 and 300 s, the critical flux has to be identified as the levels where the standard deviation of the curves starts to increase, since the value itself fluctuate, which is around 8.3 × 10^{-6} m/s and 6.1 × 10^{-6} m/s, respectively. Le Clech et al.[6] have observed a significant increase in ΔP_0 for the longer step length (7200 s), using a MBR system processing sludge and sewage, but at lower step lengths, the effect on ΔP_0 is less pronounced, which also seems to be the case with our system. Altogether, as seen in Figure 6 for our system, the critical flux determination using this parameter is very uncertain.

As seen in Figure 7, the experimentally determined critical fluxes are mostly sensitive to the used step length when the step up method is used, but also to some extent for the step up down method. For the step up method, the average critical flux value is 8.3 × 10^{-6} m/s ± 30%, whereas the value is 9.7 × 10^{-6} m/s ± 11% for the step up down method. Therefore, the results of the two methods cannot be characterized as being different within the experimental error. It should be noted that the prediction of the critical flux as 2/3 of the limiting flux, reported by Bacchin,[15] seems to be reasonable in our case although one should not really conclude much based on data with such large error. However, the "2/3" theoretical link between critical and limiting flux is based on the assumption that a flux distribution and a boundary layer thickness distribution exist along the membrane surface. Bacchin states, "the critical flux is reached when irreversible fouling occurs locally on the membrane, whereas

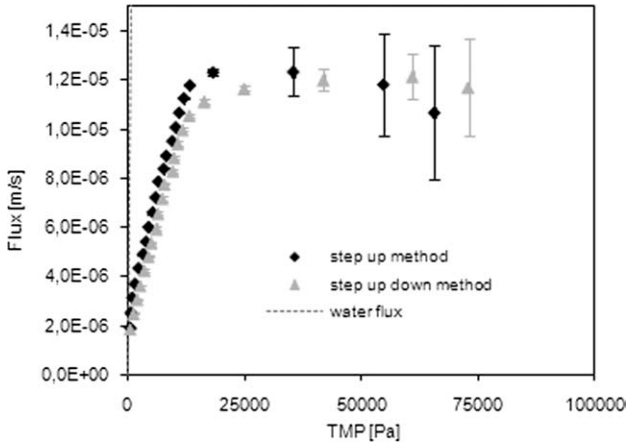

Figure 5. Flux vs. TMP for one of the *step up* experiment (step length: 1200 s, step height: 5.8 × 10^{-7} m/s, flux start level J_0: 1.9 × 10^{-6} m/s) and for one of the *step up down* experiment (step length: 600 s, step height: 5.8 × 10^{-7} m/s, flux start level: 1.9 × 10^{-6} m/s).

Standard deviation for the three identical experiments is shown with error bars and the water flux curve is shown as well.

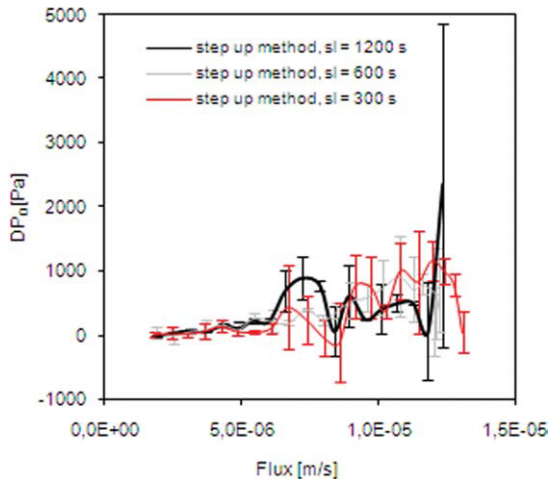

Figure 6. ΔP_0 vs. flux for the *step up* experiments. Step length: 1200, 600, and 300 s, step height: 5.8 × 10^{-7} m/s, flux start level J_0: 1.9 × 10^{-6} m/s.

Standard deviation is shown with error bars.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

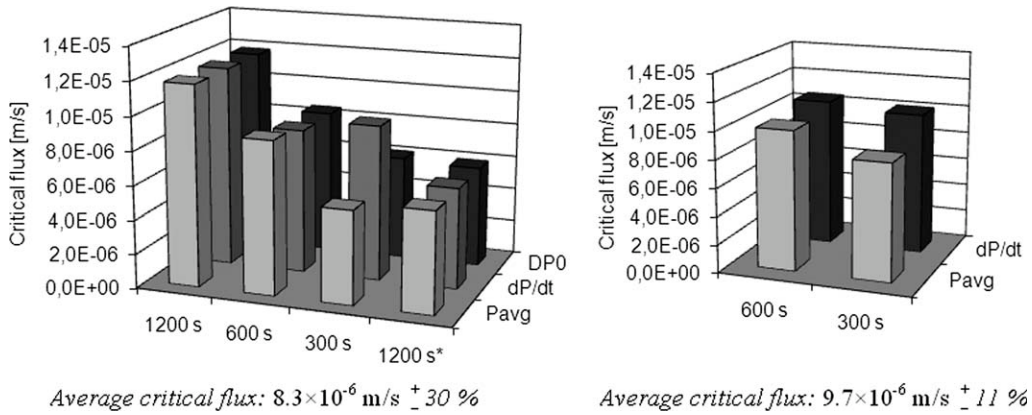

Figure 7. Critical fluxes for *step up* method (left) using step lengths of 300, 600, and 1200 s and based on different pressure related parameters (P_{avg}, dP/dt, and ΔP_0), and for the *step up down* method (right) using step lengths of 300 and 600 s using the P_{avg} and dP/dt parameters.

Step height = 5.8×10^{-7} m/s and flux start level = 1.9×10^{-6} m/s. NB: 1200 s*; experiment conducted with a step length of 1200 s, a flux start level of 4.4×10^{-6} m/s, and a step height of 1.3×10^{-6} m/s.

the limiting flux is reached when the whole membrane surface operates above the critical flux."[15] This could be the same case for our system.

Generally, the determined critical fluxes increase with increasing step length. This is opposite to the observations reported by Le Clech et al.,[6] and Choi and Dempsey[9] who reported decreasing critical fluxes with increasing step length, which might be due to the possibility for fouling to build-up during a longer step length period, causing a decreased critical flux. However, other effects must be dominant during operation of our system. Kwon and Vigneswaran,[7] and Guo et al.[8] reported no effect of different step lengths on the determined critical fluxes, which is also contradictory to our observations. Even though one has to remember that these systems are different from our system, as well as the nature of the feed solutions/suspensions are not the same, the reason and mechanism behind our observed tendency of increasing critical flux with increasing step length is not known, but part of the variation in the data might be ascribed to experimental error. Furthermore, when using step length of 1200 s, a higher flux start level of 4.4×10^{-6} m/s, and a larger step height of 1.3×10^{-6} m/s, the determined critical flux is around half the value as that with the lower flux start level and step height. However, it is surprising that less fouling is observed at longer filtration step lengths when the step height and flux start level are fixed.

Overall, it must be concluded that experimentally determined critical fluxes, in our case, are very sensitive to choice of operational parameters. Altogether, it must be stated in our case that these critical flux measurements are useless in itself as critical flux predictors. However, as will be shown later, the data will be used as starting points for verification experiments.

Fouling irreversibility

The advantage of the step up down method is, in theory, the possibility of determining the onset of irreversible fouling which by Bacchin et al.[2] is referred to as the irreversibility critical flux form. This can be done by depicting the difference in flux, TMP, and fouling rate for "same level steps," which is similar to the SWB method described by Espinasse et al.[11]

Figure 8 shows the difference in flux, TMP, and fouling rate between the same level steps, *n*. When fouling is reversible, the flux, TMP, and fouling rate difference should be zero as the pump operates at the same rate at that certain step and, therefore, an eventual polarized layer should decrease when the suction pressure is released back to a previous level. When fouling starts to get irreversible, the TMP will not be restored when moving from step *n,i* to step *n,ii* because in-between the flux has passed a higher flux step. According to Espinasse et al.,[11] a transition from a dispersed to a condensed phase occur at the membrane surface when the permeate flux is high enough to overcome the dispersive repulsive forces between the suspended components, and between the surface and the suspended components. At this stage, pore blocking or constriction is probably likely to occur. The point, where the differences in flux, TMP, and

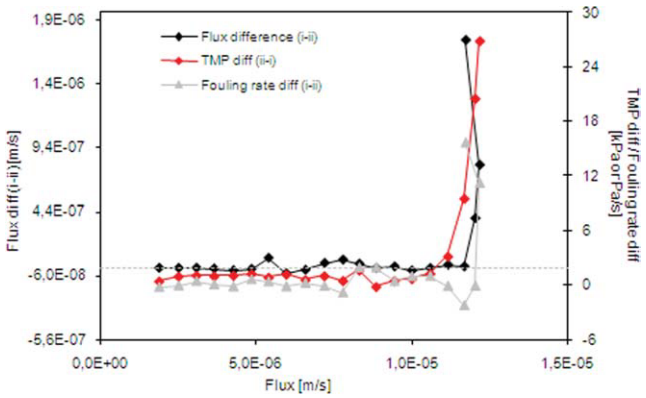

Figure 8. *Step up down* method.

Flux, TMP, and fouling rate difference between same level steps. Indication of fouling irreversibility. Step length: 600 s, step height: 5.8×10^{-7} m/s, flux start level: 1.9×10^{-6} m/s. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

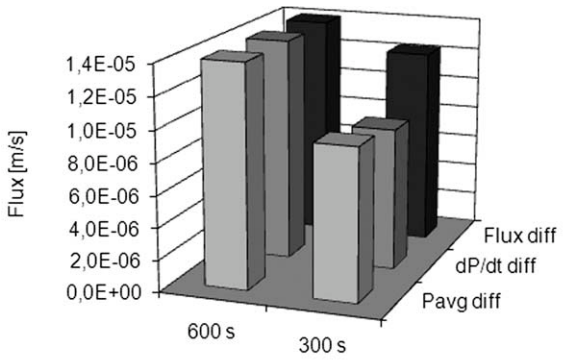

Figure 9. The flux level at the onset of irreversible fouling based on flux, *dP/dt*, and P_{avg} difference. *Step up down* method.

Step length: 300 and 600 s, step height: 5.8×10^{-7} m/s, and flux start level 1.9×10^{-6} m/s.

fouling rate are no longer zero, is taken as the irreversible critical flux form. Actually, when looking at Figure 8, it is seen that the TMP difference curve, before the break, is throughout most of the flux interval slightly above zero (0.8 kPa $\pm$ 0.4 kPa), which indicate slow build-up of irreversible fouling during the whole experiment. Therefore, it might be more correct to refer to a "sustainability of fouling irreversibility" even though 0.8 kPa $\pm$ 0.4 kPa is a very low and almost undetectable pressure difference. The flux and fouling rate difference, before the curves break, is 5.8×10^{-8} m/s $\pm$ 150% and 0.22 Pa/s $\pm$ 410%, respectively, which shows that within the standard deviation, these values are not significantly different from zero. The fluxes, at which irreversible fouling is observed (irreversible critical flux form), by using the step up down method is depicted in Figure 9.

It can be seen that the flux level, at which irreversible fouling is observed, is dependent upon the used step length as well as the method, by which the flux level is determined (flux, dP/dt, or P_{avg} difference). The main tendency, similar to the step up method, is that increased step length leads to increase in critical irreversibility flux. The average flux level for irreversible fouling is 1.1×10^{-5} m/s $\pm$ 13% which is slightly higher compared with the critical fluxes for the step up method (8.3×10^{-6} m/s $\pm$ 30%) and for the step up down method (9.7×10^{-6} m/s $\pm$ 11%). This was also expected, according to the definitions of the different types of critical fluxes given in the appendix A of Bacchin et al.,[2] as the difference between weak form of critical flux and irreversible form of critical flux is that the total hydraulic filtration resistance is the sum of membrane and adsorption resistance (weak form critical flux) and membrane, adsorption and reversible resistance (irreversible form critical flux), respectively. However, regarding the large variation in the determined critical fluxes and in the irreversible form critical fluxes, one should be very cautious in differentiating between weak form critical flux and irreversible form critical flux from our measurements as the values are all within the experimental error (standard deviation). Therefore, one cannot conclude a difference in the different determined critical flux forms.

Critical fluxes tested against constant suction experiments

As seen in the previous section, the determined critical fluxes vary quite much from 5.6×10^{-6} m/s to 1.2×10^{-5} m/s and are very dependent upon the used step length and also the combination of step height and the flux start level as well as the determination method. Therefore, the data should be verified against constant suction experiments. Three 5 and 1/2 hours constant suction experiments were conducted, each repeated three times. The three flux levels were chosen based on the average values of the determined critical fluxes from the flux-stepping experiments:

- $J_0 = 5.6 \times 10^{-6}$ m/s. This flux level was chosen in order to be below the average critical flux value.
- $J_0 = 8.3 \times 10^{-6}$ m/s. This flux level was chosen in order to seek to be around the critical flux.
- $J_0 = 1.1 \times 10^{-5}$ m/s. This flux level was chosen in order to seek to be in the supra-critical flux regime around the onset of irreversible fouling.

In Figure 10 and Figure 11 the flux and TMP versus time for the constant suction experiments are shown.

The 1.1×10^{-5}m/s experiments clearly show that this flux level is supra-critical and not sustainable. The flux in the supra-critical regime is not sustainable and decreases relatively fast in the first 4 and 1/2 hours (16,200 s), probably because of cavitations in the pump. The TMP curve at supra-critical flux increases linearly in the first 1 and 1/2 hour with a slope, fouling rate, of 14.5 Pa/s which is more than one order of magnitude larger than the acceptable fouling rate limit of 1.1 Pa/s. After around 2 hours (7200 s), the permeate pump probably started to cavitate, explaining the flux decrease and the leveling out of the TMP curve. It is seen that in the experiment at 8.3×10^{-6}m/s, the flux level might be at some critical or intermediate level with a TMP

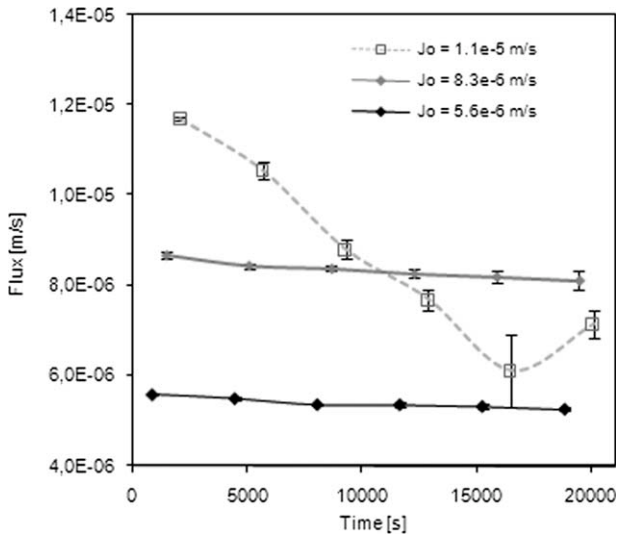

Figure 10. Flux vs. time for the constant suction experiments with flux start levels J_0 of 1.1×10^{-5}m/s, 8.3×10^{-6}m/s, and 5.6×10^{-6}m/s.

Error bars show the standard deviation between the three repetitions of each experiment.

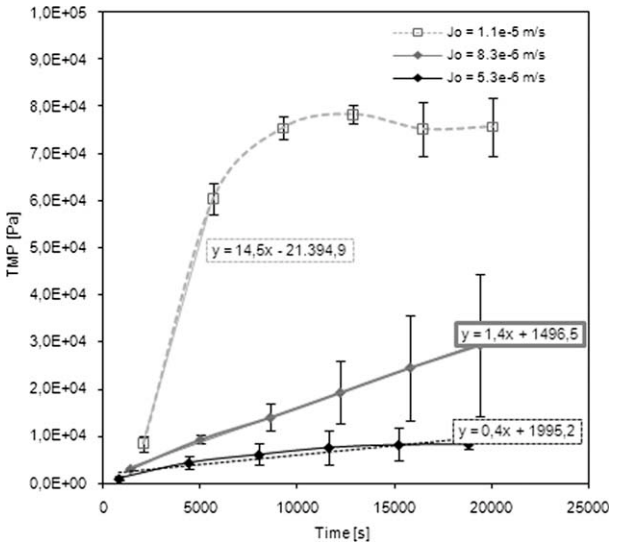

Figure 11. TMP vs. time for the constant suction experiments with flux start levels J_0 of of 1.1 × 10^{-5}m/s, 8.3 × 10^{-6}m/s, and 5.6 × 10^{-6}m/s.

Linear regression lines are shown for each of the curves. For the $J_0 = 1.1\times10^{-5}$m/s experiment, the linear regression line is only fitted to the first part of the curve until ~ $1^1/_2$ h (5400 s). Error bars show the standard deviation between the three repetitions of each experiment.

increasing almost linearly with a slope, fouling rate, of 1.4 Pa/s. This fouling rate is slightly above the earlier defined limit. Only the experiments conducted at 5.6 × 10^{-6} m/s, seems to be in a sub-critical and sustainable flux regime with a fouling rate below the accepted limit (0.4 Pa/s). The difference in the TMP curves probably indicate differences in the mode of fouling at the different flux levels or at least the degree of fouling is significantly different. Where pore blocking is probably likely to have occurred at the highest flux level, only minor depositions and pore constrictions might have occurred at the lowest flux level. However, as seen in Figure 10, the fluxes also decrease slightly in the experiments, $J_0 = 5.6\times10^{-6}$ m/s and 8.3×10^{-6} m/s, which indicate slowly, but progressively, fouling build-up on the membrane.

As mentioned earlier, step-by-step methods only determine critical fluxes for the dominant fouling species, which in our case probably corresponds to whole yeast cell, or maybe larger yeast cell debris, but still, extracellular polymeric substances (EPS) from the yeast cells can cause severe fouling and is likely also to cause part of the permeability drop. The influence of EPS on membrane fouling and on macromolecular transmission has been investigated and described in other parts of our research.[10]

Altogether, the step-by-step experiments generated critical flux values with very large standard deviation. Thus, in itself these values are useless to predict critical fluxes. However, when using these values as start guesses in constant flux verification experiments, a sub-critical and sustainable flux was actually identifiable. The duration of the verification experiment was 5 and 1/2 hours, and therefore the sustainable time period is 5 and 1/2 hours.

Conclusions

The aims of this article are

(i) to show that one cannot be sure to use a critical flux value based on one single step-by-step measurement to actually determine a critical flux.

This has been proved, as two different critical flux determination methods both generated critical flux values with large standard deviation. The average critical fluxes for the step up method is 8.3 × 10^{-6} m/s ± 30%, whereas the average of the critical fluxes for the step up down method is 9.7 × 10^{-6} m/s ± 11%. Thus, the two methods gave the same results within the experiment error, and because of the large standard deviation, the critical flux values are in itself useless as predictors for sustainable flux regimes. Furthermore, the theoretical possibility of determining the onset of irreversible fouling, using the step up down procedure, was not achievable in our case, and differentiation between irreversible critical flux form and weak critical flux was, therefore, not possible because of the large standard deviation in the obtained values. How to use the two flux-stepping procedures have been described in detail.

(ii) to show that step-by-step determined critical fluxes has to be verified against constant flux experiments.

Using the step-by-step determined critical fluxes as start guesses for constant flux verification experiments, a sub-critical and sustainable flux was actually identified. Distinctions in the fouling levels were observed at flux levels of 1.1 × 10^{-5} m/s, 8.3 × 10^{-6} m/s, and 5.6 × 10^{-6} m/s. Only the 5.6 × 10^{-6} m/s level was in the sub-critical flux region and sustainable. Thus, the lowest determined critical flux values turned out to be at a sustainable level.

(iii) to emphasize that when talking about critical fluxes and sustainable flux regimes, a time period for sustainability has to be assessed.

The duration of the constant flux verification experiments was 5 and 1/2 hours. Thus, in our case the sustainability time is 5 and 1/2 hours. Whether this flux level is sustainable after 5 and 1/2 hours cannot be assessed from these data.

Overall, one should be very cautious in talking about sustainable fluxes based on critical flux values determined from step-by-step determination procedures. One should at least consider the three conclusions of this article in the evaluation of critical flux results obtained from flux-stepping experiments.

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