

Perspective of Critical Flux on Membrane Filtration

Maneerat Tiranuntakul¹

บทคัดย่อ

บทความนี้นำเสนอความรู้หลักที่เกี่ยวกับฟลักซ์วิกฤตและการกรองของเมมเบรน โดยในส่วนแรกของบทความได้นำถึงความหมายของฟลักซ์วิกฤตทั้งแบบประเภทสตรองและแบบวีค ในส่วนถัดมาได้แสดงรายละเอียดของการหาค่าฟลักซ์วิกฤตซึ่งอยู่บนพื้นฐานของหลักเกณฑ์และหลักการวิเคราะห์ที่แตกต่างกันสามรูปแบบ อันได้แก่ การใช้เทคนิคสมดุลมวล การสังเกตโดยตรง และการติดตามแนวโน้มการกรอง เป็นต้น นอกจากนี้ยังได้กล่าวถึงความสัมพันธ์ระหว่างค่าฟลักซ์วิกฤตและปัจจัยที่มีอิทธิพล เช่น คุณสมบัติของสารที่ทำการกรอง คุณสมบัติของเมมเบรน และลักษณะของการไหล เป็นต้น และเนื่องจากยังไม่ได้มีการกำหนดรูปแบบมาตรฐานของการหาค่าฟลักซ์วิกฤต ดังนั้นบทความนี้จึงได้กล่าวถึงการศึกษาเปรียบเทียบอิทธิพลการหาค่าฟลักซ์วิกฤตในรูปแบบต่างๆ ไว้ด้วย ในส่วนท้ายสุดบทความนี้ได้ต่อยอดความเข้าใจในปัญหาของกรณีการอุดตันภายใต้สภาวะการดำเนินงานที่ต่ำกว่าค่าฟลักซ์วิกฤต โดยได้อธิบายรวมถึงปรากฏการณ์ฟลักซ์วิกฤตเฉพาะที่และเสนอแนะให้ใช้การทำความสะอาดเมมเบรนเป็นระยะเพื่อแก้ปัญหาด้วย กล่าวโดยสรุปคือบทความนี้ตระหนักถึงความเชื่อมโยงระหว่าง ฟลักซ์วิกฤต ปัจจัยที่มีอิทธิพล และการอุดตันของเมมเบรน ซึ่งอาจเป็นประโยชน์ต่อผู้อ่านในด้านการปรับปรุงการดำเนินงานของระบบการกรองด้วยเมมเบรนต่อไป

คำสำคัญ: ฟลักซ์วิกฤต การกรองด้วยเมมเบรน การอุดตัน

Abstract

This paper aims to deliver the principle knowledge about critical flux and membrane filtration. Firstly, definitions of critical flux are introduced in both strong form and weak form. Determinations of the critical flux are mentioned in three categories based on differences of considerations and analysis techniques including material balance technique, direct observation and filtration profile monitoring. In addition, correlations between critical flux values and affecting factors are also focused e.g. feed properties, membrane properties and hydrodynamic properties. Due to no standard protocol for critical flux evaluation, comparison studies of critical flux under various influencing assessments are presented. In the last section, to clarify the case of sub-critical flux fouling problem, the local critical flux phenomenon is explained and the membrane cleaning periodically is suggested. In summary, this paper, perhaps, makes more aware of the complicated links among critical flux, influencing parameters and membrane fouling which might be more useful to all readers for improving of membrane application.

¹ Lecturer, Chemical Engineering Division, Faculty of Engineering, Rajamangala University of Technology Krungthep, Tel. 08-3389-2469, E-mail: maneerat.t@rmutk.ac.th

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1. Introduction

Membrane filtration systems have been developed for more than 150 years, starting from preliminary research and development through to modern widespread use. By the 1960s, elements of modern membrane science had been developed and used in laboratories. By the 1980s, the problem of slow permeation rates and the issue of packaging a large membrane surface area into low cost modules had been overcome.

However, membrane filtration process still has a major disadvantage of membrane fouling which affects on operational and energy costs for membrane replacement, membrane maintenance and membrane cleaning. A suggested border to handle this fouling problem called critical flux was introduced just last decades ago. Thus far, significant differences of critical flux based on definitions, determination methods, influencing factors and state of fouling have not been summarized. Therefore, this paper aims to broaden a principle knowledge based on critical flux and membrane filtration which might be more useful to all readers who involving in membrane application.

2. Concept of Critical Flux

In membrane processes, permeate flux is an important parameter for determining fouling rate [1]. It is generally considered that higher productivity can be achieved by operating at a higher flux, which may initiate more fouling profoundly. In order to prolong the membrane life and make a compromise between high production rate and low fouling rate, the so-called critical flux is applied. It has been reported that fouling

is not observed when the flux was kept or maintained below the some certain flux (or critical flux) [2].

The concept of critical flux was introduced in 1995. The postulation of the critical flux was based on the following definition [3]: “The critical flux hypothesis for microfiltration is that on start-up there exists a flux below which a decline of flux with time does not occur; above it, fouling is observed. This flux is the critical flux and its value depends on hydrodynamics and probably other variables.”

Critical flux was defined again another way based on the flux below which there is no deposition of colloids on the membrane [4]. In general, these will not give the same flux value. Beyond the critical flux, irreversible fouling of suspended solids forms a stagnant, consolidated and aggregated layer on the membrane surface, which can make flux decline rapidly. On the other hand, below the critical flux condition, called sub-critical flux, it has been reported that fouling is not observed [2]. Consequently, the concept of critical flux is a key parameter for characterizing fouling. Summary of all critical definitions is shown in table 1 [5].

Table 1 Definitions of critical flux [5]

Definitions	Discrimination	Determination based on
Strong form of critical flux	No fouling	Filtration profile compared with water flux linearity
Weak form of critical flux	Fouling independent of solvent transfer	Linearity of flux-TMP
Critical flux for reversibility	Reversible fouling	Irreversibility

There are two forms of critical flux: strong form and weak form. The strong form states that the sub-critical flux and TMP relationship shows a straight

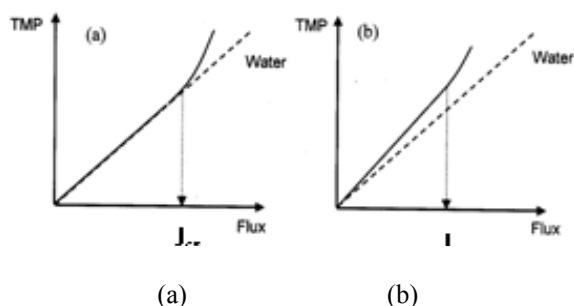


Figure 1 Schematic diagrams of (a) the strong form critical flux, and (b) the weak form critical flux [6]:

line of the same slope as that of pure water for the same operating pressure, while the weak form shows a straight line, the slope of which differs from that of pure water. Any deviations from the straight line for both forms indicate above critical flux conditions. Figure 1 shows the strong and weak forms of the critical flux.

3. Determination of Critical Flux

3.1 Based on Material Balance

Material balance technique was employed by measuring the rate of particle deposition at the membrane surface and observing the variation of particle concentration in the feed entering and leaving the modules. The defined critical flux was the maximum flux at which the feed concentration did not change. The critical flux for latex particles determined by mass balance and by TMP monitoring was studied and found that the critical fluxes identified by resistance assessment were significantly higher than those based on mass balance [7]. The difference between these two critical fluxes increases with the particle size. This indicates that the assessment of filtration resistance cannot always determine the flux

at which particles start to deposit onto the membrane if the particles can form a loose cake layer.

3.2 Based on Direct Observation

Determination of critical flux based on non-intrusive observation was experimented by Fane and teamwork [8]. They studied the critical flux of yeast and latex particles in cross-flow microfiltration using in situ observation of particle deposition on the membrane surface through a microscope and video camera recording. The critical flux was recognized as the highest flux where the particle deposition on the membrane surface remained unobservable. The particle build-up became significant as the flux exceeded the critical flux. Relying on light transmission through the system, this method requires a transparent membrane and this viewing technique is restricted to particles greater than 0.5 μm due to the magnification of the microscopic lens [9]. This method was used to identify critical flux for filtration of yeast, latex and bacterial particles [10].

3.3 Based on Filtration Profile

In general, if the filtration is performed at a fixed permeate flux, the increased TMP is monitored during filtration. An increase of TMP is due to the increase in filtration resistance. In this case, the critical flux can be defined as the highest flux where the filtration resistance remains constant (also constant TMP). This determination method can be performed with long term filtration or conveniently with shorter filtration duration using the flux stepping technique. The flux incremental technique has been widely used in many MBR researches [11]-[14]. In this method, the fixed flux filtration is carried out for a certain time and this procedure is repeated by incrementally

increasing the flux until a noticeable steady increase in trans-membrane pressure is observed.

In comparison, determinations of critical flux based on material balance and direct observation are suitable for lab scale systems due to its complicated detection and these two methods can predict a critical flux in a very first state even no changing of filtration profile. In the contrary, only determination of critical flux based on filtration profile is applicable for full scale systems due to its simplicity and feasibility.

4. Factors Affecting the Critical Flux

4.1 Feed Properties

1. Particle Size

Several sizes of latex particles ranging from 0.1 to 11.9 μm were tested in a crossflow micro-filtration system [7]. For small particles from 0.1 to 0.46 μm , the critical flux decreased as the particles size increased due to the greater Brownian back diffusion force of the smaller particle size. The reverse trend occurred in larger particles from 0.46 to 11.9 μm , which might be due to the cake formed by the large particles being too loose to create resistance compared with the membrane resistance. This phenomenon can be explained using the concentration polarization (CP) model. In this CP model, the sub-micron particles are likely to cause Brownian diffusivity while shear induced hydrodynamic seems to be dominant for micron sized particles.

2. Feed Concentration

Some studies [6],[7] reported the decrease of critical flux with the increase of particle concentration due to the higher particle deposition on the membrane at the higher concentration. On the other hand, the increase in MLSS concentration to 12 g/l could noticeably reduce the fouling performance [14]. In summary, the effect of MLSS concentration on

the critical flux is not very obvious and difficult to compare results from different researches due to the sludge complexity.

3. pH and Ionic Strength

It was observed that ionic strength of particles has a significant effect on the critical flux [15]. The increase in ionic strength from 10-5-10-2 M decreased the critical flux values, thereafter the critical flux increased. Also, a considering surface charge of the particles as one of the influencing factors for particle back-transport mechanisms was suggested [16]. The effects of pH on critical flux presented that the critical flux at pH 4.8 was found lower than at pH 3.0 and pH 9.0 [17].

4.2 Membrane Properties

1. Membrane Pore Size

The effect of membrane pore size on the critical flux using 50 kDa and 100 kDa membranes for 5% baker's yeast filtration showed that the larger membrane pore size tended to have lower critical flux due to the internal fouling of cell debris and small components [18]. The critical flux of 0.4% BSA solution was increased with tracked-etched membrane pore sizes (0.1, 0.2 and 0.4 μm) [19]. On the other hand, the critical flux was insensitive to some membrane pore sizes (0.1, 0.2, 0.45 and 0.65 μm) [7]. This phenomenon can be explained in that the total drag force for different membrane pore sizes was identical at the same flux and causing similar deposit latex particles.

2. Zeta Potential

Many studies found that neither the zeta potential of the silica particles (0.53 μm) nor the membrane had an impact on the critical flux. Some earlier works reported that the specific resistance is strongly dependent on zeta potential of colloids [20],[21]. The zeta potentials changed significantly after fouling by

humate reducing cationic functionality and adsorption of anions resulting in reduced zeta potentials [22]. The modification of membrane surfaces by oxidation increased the fouling and mitigating fouling by decreasing of interaction between foulants and membrane zeta-potential [23].

3. Hydrophilicity/hydrophobicity

The effects of hydrophilic and hydrophobic membranes on the critical flux was compared and found that the larger critical flux appeared for hydrophilic membrane [24]. Protein could deposit above and below the apparent critical flux using the hydrophobic membrane while the coverage was only found above critical flux when the hydrophilic membrane was used [25].

4.3 Hydrodynamics

Some hydrodynamic factors such as cross-flow velocity and air sparging have been accounted to affect the critical flux. Most researches showed the increase of critical flux with increase of cross-flow velocity and air sparging [8], [26] due to shear-induced diffusion and inertial lift.

In addition, the effects of the constant pressure operation and constant flux operation on the critical flux was compared [27],[28]. The results showed that the constant flux operation created less hydraulic resistance than constant pressure operation due to different fouling histories and fouling initiation. A linear relationship between air flow rate and critical flux in the submerged hollow fibre system was reported [26].

5. Comparison of Critical Flux Assessment

Until now, there is no standard methodology or precisely agreed-upon protocol to define the exact value of the weak form of critical flux. The critical fluxes evaluated by different methods were compared and plotted in Figure 2 [32]. Obviously, the critical

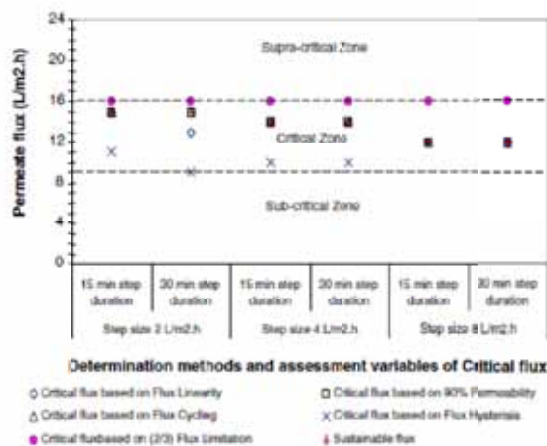


Figure 2 Comparison of critical fluxes obtained from different variables and determination methods [32] critical flux.

fluxes were affected by the filtration variables, namely step height and step length. The decline of critical flux as the step height increases has been noticed in most determination methods namely; flux linearity, 90% permeability, and reversibility of flux cycling, which is similar to the observation based on flux linearity. Of course, it would be better to use small step heights to determine critical flux values in order to prevent large errors from flux averaging. Unlike other methods, hysteresis of flux stepping filtration shows a positive relationship between the step height and critical flux values. On the other hand, the step length has no obvious effect on the critical fluxes assessed from all determination methods. Noticeably, only the critical fluxes based on the two-third limiting flux method are constant and independent from the influence of step height and step length [33].

In fact, fouling was reported to develop in the transient behavior [34]. Therefore, it is reasonable that various determination methods of critical flux can give slightly variations of the critical flux values. From Figure 2, the highest and lowest critical flux values are obtained from the two-third limiting flux

and the hysteresis of stepping filtration, respectively. It is suggested that the two-third limiting flux and the flux hysteresis are likely to indicate upper and lower border of the critical zone, respectively. Operating beyond this point is likely deleterious to membrane operation. A short term experiment is only sufficient to indicate critical flux but not relevant to the stability of this critical flux over longer periods of time [34]. Therefore, the recommended flux for membrane operation is dependent upon the application period which some fouling rate can be tolerated when operating in the short term, but unacceptable for long term filtration.

6. Sub-critical Flux Fouling in the Long Term Run

Based on the critical flux concept, operation of the membrane process under sub critical flux is generally recommended due to avoiding of fouling problem. However, the development of the fouling process can happen in the long term operation despite the choice of sub-critical conditions. It is based on the following phenomenon (Figure 3): during the first period, solute-membrane interactions provoke a reduction in the number of pores open to the filtrate flow. As the permeate flow is held constant during the experimental run, this reduction of the area open to the flow is expressed a gradual increase in circulation rate, or local flux J_p , in the pores remaining open. In the absence of regular membrane regeneration, the increase slowly intensifies as the pores close, and may lead to the local flux reaching a level equal to the critical flux. A deposit then forms on the membrane, translating to very high hydraulic resistance: this marks the onset of the second filtration period and the cake fouling has been occurred [35]. This local critical flux phenomenon confirmed the importance of membrane cleaning periodically in the long term run of any membrane systems.

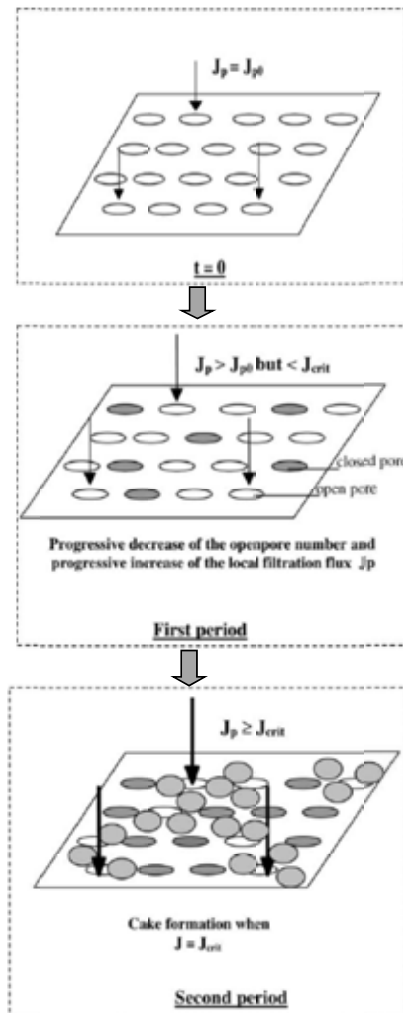


Figure 3 Changes of filtration area and permeate local flux and consequences on fouling mechanism.

7. Conclusions

The critical flux concept is useful in understanding and improving the operation of membrane filtration systems. Critical flux can be categorized as a strong form and a weak form. Determinations of the critical flux have been proposed in three categories based on differences of considerations and analysis techniques including material balance technique, direct

observation and filtration profile monitoring. Critical flux can be affected by three types of factors namely feed properties, membrane properties and hydro-dynamics. Up to date, assessment parameters on critical flux were studied and indicated that the critical flux decline as the step height increased regardless of step length influence and smaller step height are recommended due to avoiding large error of flux averaging. Moreover, the development of the fouling process can happen in the long term run despite the choice of sub-critical flux operation due to local critical flux phenomenon. Therefore, membrane cleaning periodically in the long term run is suggested. In summary, this paper, perhaps, makes us become more aware on the complicated links among nature of critical flux, influencing parameters and membrane fouling which might be more useful to all readers involving in membrane application.

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