CHAPTER II

LITERATURE REVIEW

Membrane filtration is a technology with wide interests as it can be used in product and process developments. Nowadays, the membrane filtration technology, especially microfiltration, has been applied in several aspects in food industries.

2.1 Membrane filtration processes in food industry

Membrane filtration is a solid-liquid separation process. The liquid and insoluble solids which are smaller than membrane pore pass through a porous membrane, while membrane retains the solid particles which are larger than membrane pores. The suspension is known as the feed slurry, the liquid component that passes through the membrane is called the filtrate (or permeate) and the membrane itself is referred to as the filter medium. The separated solids are known as the filter cake, once they form a detectable layer covering the upstream surface of the medium. However, the solid particles which are smaller than membrane pore can be retained within the membrane pore and/or the filter cake. The flow of permeate may be brought about by the application of a pressure greater than atmospheric upstream of the membrane (Brennan *et al.*, 1976).

The membrane filtration has been applied in the food industry, such as separation of solid and liquid components, clarification for removing small quantities of insoluble solid from a valuable liquid. To removal of very fine particle as well as microorganism from liquid foods, the membrane pore size between 0.2-10 μm is used

and the process is called microfiltration. In filtrations, a cake may build up on the membrane, where the amount of solids is very small, they may become enmeshed within the structure of the membrane (Brennan *et al.*, 1976).

The microfiltration has been used in food industries to clarify food, improve stability and eliminate small colloidal particles and microorganisms causing food spoilage without apply heating, especially in liquor industries such as wines and beers, so called cold sterilization. Thus, the flavor of products was maintained without any loss during the process. Moreover, it can also extend shelf life of products. Presently, applications of membrane technology are found in many food industries as the followings;

- Separation: To separate solutes and small particles from solutions, e.g., separation of whey proteins from milks (Vandenhorst and Hanemaaijer, 1990).
- Concentration: To separate solutions or solvents that can pass through membranes so that solutes are concentrated in the sample, e.g., concentration of fruit juices (Jonsson and Tragardh, 1990).
- Purification: To extract high value substances to be more purify and recover some substances back, e.g., extraction of flavor substances from fruit juices, extraction of proteins and amino acids from wastewater (Jaouen and Quemeneur, 1992).
- Clarification: To extract some substances and particles causing turbidity and colloidal instability in foods, e.g., clarification of beer and wine products. (Burrell and Reed, 1994; Gan *et al.*, 1997; Gan *et al*, 2001; Fillaudeau, and Carrere, 2002), production of mineral water (Vanderhorst and Hanemaaijer, 1990).

- Cold-pasteurization and cold-sterilizations: To remove microorganisms that can cause food spoilage from food products without heating, so food products can be clear and maintain flavor, and the shelf life can also be extended, e.g., beer productions (Gan *et al.*, 2001), wine and vinegar productions (Jonsson and Tragardh, 1990), milk productions (Vandenhorst and Hanemaaijer, 1990).

2.2 Microfiltration theory

Microfiltration is defined as the process that uses the pressure-driven flow through a membrane is used to separate the particles having size 0.2-10 μm from fluids (Toledo, 1991). The particles within this size range include large soluble macromolecules and microorganisms. Usually the pressure drop used across the membranes varies from 1 psi to 50 psi. Types of membranes are extremely varied and can be ceramics, polymers, and so on. Much different geometry of membranes is used. These include spiral-wound, plate and frame, hollow fiber, cartridge filters with pleated membranes (Geankoplis, 2003).

In filtration, a cake is formed on the porous surface of the membrane and the process is known as cake filtration. In the initial stage of filtration, the first particles of solid to encounter the membrane become enmeshed in it, reducing its open surface area and increasing the resistance it offers to the flow of permetae. As filtration proceeds a layer of solids builds up on the surface of filter and this layer, or cake, increases in thickness with time. Once formed, this cake becomes the primary filtering medium. Permeate passing through a filter encounters three types of resistance, namely (a) that offered by the channels and ports of the filter itself, (b) that

offered by the filter medium, and (c) that offered by the filter cake. The total pressure drop across the filter is equivalent to the sum of the pressure drops resulting from these three resistances. Usually, the pressure drop through the channels and a port of the filter itself is neglected in calculations (Brennan *et al.*, 1976).

2.2.1 Filtration resistance

An important property of a membrane is permeate flux or flux, which is defined as the rate of permeate volume (or mass) passing through a unit area of membrane and can be expressed as Equation 2.1 (Fane, 1984; Baker *et al.*, 1985).

$$J = (1/A)\, dV/dt \qquad (2.1)$$

where J = permeate flux (m/s)

V = volume of permeate (m^3)

t = filtration time (s)

A = membrane area (m^2)

The ΔP is pressure difference over a membrane which is the difference between the pressure at the feed side and the pressure at the permeate side. The relationship between J and ΔP is defined by a modified form of Darcy's law which is expressed as Equation 2.2 (Fane, 1984; Baker *et al.*, 1985; Lee *et al.*, 2000; Lee *et al.*, 2005).

$$J = \Delta P/\mu (R_m+R_c) = \Delta P/\mu R_t \quad (2.2)$$

where ΔP = transmembrane pressure (Pa)

μ = viscosity of permeate (Pa·s)

R_m = membrane resistance (m^{-1})

R_c = cake resistance (m^{-1})

R_t = total resistance (m^{-1})

The cake resistance is proportional to the mass of cakes deposited on the membrane surface, can be expressed as Equations 2.3 and 2.4:

$$R_c = \alpha\, m\, /A \quad (2.3)$$

$$R_c = \alpha VC/A \quad (2.4)$$

where α = specific cake resistance (m/kg)

m = mass of cake (kg)

A = membrane area (m^2)

V = volume of permeate (m^3)

C = concentration of suspended solids (kg of suspended solids /100 kg feed)

During membrane filtration, some constituents of the feed that is larger than membrane pore size deposit on the membrane surface and/or in the membrane which are the components that are smaller than the membrane pore size. This retention process is often referred to as fouling of the membrane and causes a decrease of the flux. The easily removable part of the retained material is called the reversible fouling layer, the remaining part is called the irreversible fouling layer. The feed constituents that are retained on or in the membrane surface are called foulants. The retention of feed constituents causes an increase of the total resistance over the membrane, resulting at a constant ΔP in a decreased flux. The decrease in flux that is found during membrane filtration is schematically drawn in Figure 2.1.

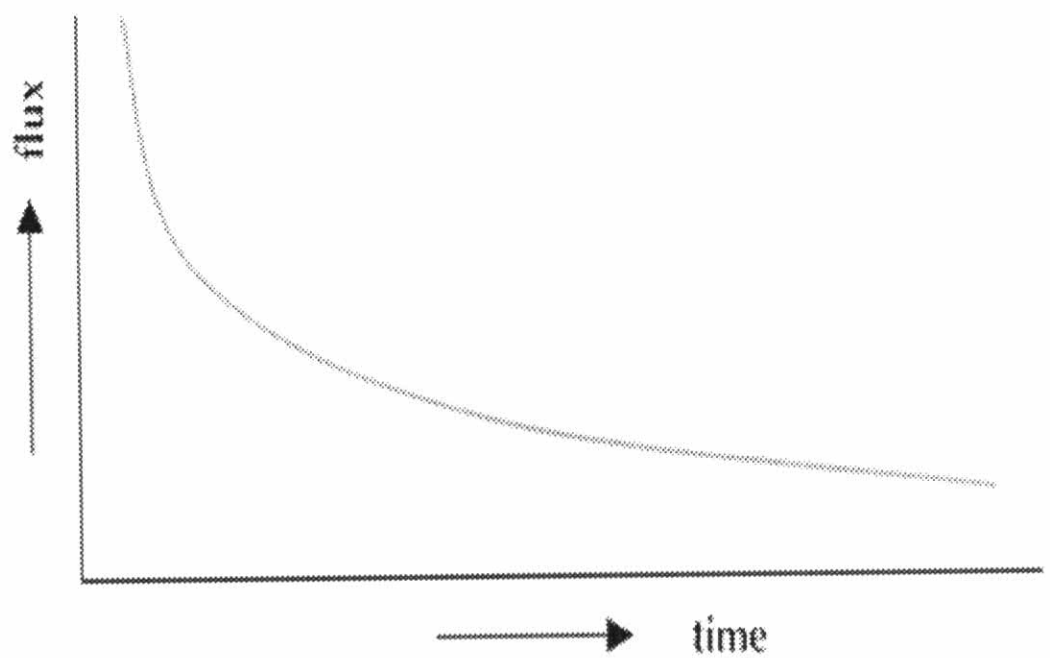

Figure 2.1 Flux decline with time due to fouling of feed during microfiltration at constant a pressure.

Feed constituents that are retained can be found on several places near the membrane surface. Essentially, five so-called 'fouling mechanisms' can be distinguished; each mechanism contributes to the total resistance over the membrane. Fouling is caused by the deposition of suspended solids and colloids, organic and inorganic foulants and biological foulants inside and on the membrane. The extent to which the membrane fouls depends on the type of membrane and the constituents in feed suspension. The total resistance to flux, is caused by several factors (Mulder, 1991; Bowen, Calvo, and Hernandez, 1995; Blanpain and Lalande, 1997):

- Membrane reistance: Membrane resistance depends on the membrane thickness, nominal pore size, pore density/porosity, pore size distribution, pore depth and tortuosity.
- Resistance due to internal colloidal foulants.
- Resistance due to the formation of highly concentrated layer adjacent to the membrane, concentration polarization. Concentration polarization: is the accumulation at the upstream surface of the membrane, of solute which are rejected or retained by the course of microfiltration.
- Resistance due to the formation of gel layer which due to the increasing concentration of particles near the surface of the membrane.
- Resistance due to pore-blocking.

2.2.2 Cake filtration theory

In cake filtration, permeate flow through the pores in the cake is dependent upon the pressure differential across the cake and the resistance to flow. The total resistance increases with increasing cake thickness; thus, permeate flow decreases with time of filtration. The total pressure drop across the filter is the sum of the pressure drops across the membrane and that across the cake.

At any permeate flux, the resistance of the membrane and the resistance of the cake can be expressed as following equations (Toledo, 1991).

$$R_m = \frac{\Delta P_m}{\mu J} \qquad (2.5)$$

where ΔP_m = the pressure drop across the membrane (Pa)

$$R_c = \alpha\,(m/A) = \frac{\Delta P_c}{\mu J} \qquad (2.6)$$

where ΔP_c = the pressure drop across the the cake (Pa)

α = specific cake resistance (m/kg)

Since: $$\Delta P = \Delta P_m + \Delta P_c \qquad (2.7)$$

Therefore: $$\Delta P = J\left(\mu \alpha \frac{m}{A} + \mu R_m\right) \qquad (2.8)$$

The mass of cake approximately equals to the product of permeate volume and concentration of suspended solids.

$$m \approx VC \tag{2.9}$$

$$J = (1/A)\, dV/dt \tag{2.10}$$

Therefore, Equation 2.8 can be expressed as:

$$\Delta P = \frac{1}{A}\frac{dV}{dt}\left(\frac{\mu\alpha CV}{A} + \mu R_m\right) \tag{2.11}$$

$$\frac{1}{AdV}\frac{dt}{} = \frac{\Delta P}{\frac{\mu\alpha CV}{A} + \mu R_m} \tag{2.12}$$

Equations 2.10 in Equations 2.12 give:

$$1/J = \frac{\mu\alpha C}{A\Delta P}V + \frac{\mu R_m}{\Delta P} \tag{2.13}$$

From Equations 2.13, it is possible to determine the α from filtration data. The 1/J are plotted against permeate volume (V), the slope of the slope of 1/J vs V is $\mu\alpha C/A\Delta P$, respectively. The intercept on the ordinate at V = 0 equals to $\mu R_m/\Delta P$ (Chudacek and Fane, 1984; Toledo, 1991; Foley, 2006).

Therefore, α can be calculated from slope of the plot between 1/J vs. V as Equation 2.14.

$$\alpha = \frac{s A \triangle P}{\mu C} \qquad (2.14)$$

where s = slope of the plot between 1/J vs. V

2.2.3 Cake compressibility

Compressibility of the cake layer can be modeled as a variation in the specific cake resistance as a power law function of the pressure as follows (Chudacek and Fane, 1984; Tanaka *et al.*, 1994; McCarthy *et al.*, 1998; Lee *at al.*, 2000):

$$\alpha = \alpha_0 (\Delta P)^n \qquad (2.15)$$

where α_0 = specific cake resistance at reference pressure (m/kg)

n = cake compressibility

The α related primary to the size and shape of the particles forming the cake, and n is the cake compressibility. For n = 0, the cake is incompressible cake; for n > 0 the cake is compressible cake. Compressible cake exhibit a decrease in porosity void volume, compactness and an increase in the α as a compressive pressure is increased. When the value n = 1, the cake is completely compressible. The permeate flux is independent of ΔP. Compressible cakes can be seen in microfiltration of organic materials, especially the cake formed of microorganisms as previous reported by Nakanishi, *et al.* (1987), Tanaka *et al.* (1994, 2001), McCarthy *et al.* (1998), and Machathy, Walsh, and Foley (2000a, 2000b). When a cake is

incompressible, its porosity and resistance are independent of the imposed pressure drop.

The α can be related to the cake porosity, morphology of the particle, and particle size using Carman-Kozeny equation (Fane, 1984; Foley, 2006) as follows:

$$\alpha = K (1 - \varepsilon) S^2 \delta / \varepsilon^3 \rho_s \qquad (2.16)$$

where K = Kozeny constant

ε = porosity of the cake

S = solid surface area per unit volume of solids (m^2/m^3)

δ = cake thickness (m)

ρ_s = density of the particles comprising the cake (kg/m^3)

For rigid spherical particles of radius (r), the specific surface area is $S = 3/r$, the porosity of a randomly packed cake is approximate 0.4, and the constant K is approximate 5 (Lee *et al*, 2003; Foley, 2006).

Generally, the cake resistance is proportional to the cake thickness, the α per unit thickness can be expressed as:

$$\alpha_1 = R_c / \delta \qquad (2.17)$$

where α_1 = α per unit thickness (kg^{-1})

Also, the α on mass basis can be expressed as:

$$\alpha = R_c / w \qquad (2.18)$$

where w = mass of cake deposited per unit area of membrane (kg/m^2)

These quantities can be related to porosity of the cake and density of the particles as follows:

$$w = \rho_s (1-\varepsilon) \delta \qquad (2.19)$$

$$\alpha_1 = \rho_s (1-\varepsilon) \alpha \qquad (2.20)$$

From the Carman-Kozeny equation, Endo and Alonso (2001) developed the equation which α was expressed as a function of particle shape, size distribution, porosity, and particle density as following.

$$\alpha = (180/\rho_s D_{vg}^2) ((1-\varepsilon)/\varepsilon^3) (\kappa / \exp (4 \ln^2 \sigma_g)) \qquad (2.21)$$

where D_{vg} = geometric mean diameter of volume equivalent diameter on a number basis

σ_g = geometric mean standard deviation

κ = dynamic shape factor

2.3 Factors affecting microfiltration performance

The parameters affect filtration efficiency of the microfiltration process is membrane characteristics, feed characteristics, and operating characteristics. Microfiltration performance is the most important part of microfiltration. The characteristic of membrane which are morphology (pore size and numbers of pore per unit area of membrane) and chemical properties affect microfiltration performance (selectivity and productivity). Selectivity is expressed as retention or separation factor in the unit of $L/m^2.h$. The following section is dealing with the research results on effects of microfiltration performance.

2.3.1 Membrane characteristics

2.3.1.1 Chemical properties

The main chemical properties of membrane are hydrophilicity and hydrophobicity which depend on material composition of membrane. In an aqueous environment, membrane has an attractive repulsive interaction to water. The material composition of membrane and its surface chemistry determine its interaction with water. A hydrophilic membrane has a high surface tension and the ability to form hydrogen bonds with water (Marshall, Munro, and Tragard, 1993; Persson *et al.*, 1993).

In aqueous media, particle that foul are likely to be hydrophobic. Generally, hydrophobic particles prefer to attach to any material including membranes which less hydrophilic than water. Upon fouling, flux decreased. To prevent fouling,

the membrane requires a surface chemistry that prefers binding to water over alter material. Therefore, to filter aqueous media hydrophilic membrane should be selected over hydrophobic membrane. The following research results support this statement. Metsamuranen, Howell, and Nystrom (2002) reported that the filtration of yeast suspensions using hydrophobic polysulphone membranes had lower flux than that with hydrophilic cellulose membranes due to hydrophobic interactions between the membrane surface and the compositions of yeast suspensions resulting in a better adsorption at the membrane surface and a higher fouling formation.

2.3.1.2 Membrane pore size

The pore size of membrane is another factor controlling fouling and affecting the rejection of feed compositions both soluble compounds and suspended solids. If the pore size is smaller than the size of particles or molecules of components in the feed stream, then it can affect a formation of fouling on the membrane's surface, which is called a surface fouling. If the pore size is larger than the size of particles or molecules of components in the feed stream, then these components can transport into the pores causing fouling inside the pore, which is called a pore fouling. The higher fouling formation results in the smaller pore size of the membrane, and thus causes a higher filtration resistance and a lower permeate flux. (Filladeau and Carrere, 2002)

Kawakatsu *et al.* (1993) found that the filtration of yeast suspensions having yeast particles of approximately 3-4 μm has the least steady state flux when using membranes with the pore size less than 0.5 μm. For the filtration with the pore size smaller or larger than 0.5 μm, the steady state flux was higher than that with

0.5 μm pore size, and the resistance was high due to the clogging inside the pores and the filtration resistance from high surface fouling. The filtration using membranes with large pore sizes will have higher permeate flux than that with small pore sizes, as found in the filtration of rough beer that the filtration with 1.3 μm pore size membranes had higher permeate flux than those with 1.0 and 0.5 μm pore size membranes, respectively. (Burrell and Reed, 1994).

Fukomoto, Delaquis, and Girard (1998) studied the filtration of apple juices and found that the filtration using a ceramic membrane with the pore size of 0.02 μm had higher permeate flux than that using a membrane with the pore size of 0.2 μm. Since the apple juices after depectinisation would have a high concentration of small suspended solid particles around 0.1-0.5 μm causing in the clogging on the membrane with the pore size of 0.2 μm and resulting in a high membrane fouling and a low permeate flux. Furthermore, Jaffrin, Gupta, and Chaibi (1993) observed similar results that the filtration of red wines using membranes with 0.4 and 0.8 μm pore sizes had higher permeate flux than that using membranes with 1.5 and 3 μm pore sizes, respectively.

Selection of membranes with suitable pore sizes will give sufficient efficiency in removal of microorganisms in food. Hence, it can be considered that a filtration with suitable membranes will be an alternative method as a cold sterilization for food. Besides, the selection of membranes with unsuitable pore sizes will cause a rejection of components in the feed stream so that they can pass through membranes along with a large quantity of permeate resulting in poor quality of permeate. Burrell and Reed (1994) studied the filtration of beer samples and found that permetae was clear as required when using membranes with the pore size of 0.5 μm and was cloudy when using membranes with large pore size of 1.3 μm due to large particles including

yeast passing through the membrane. Herath *et al.* (2000) found that the filtration of samples with microorganism of *Pseudomonas diminuta* bacteria at 1×10^7 CFU/cm^2 using polyethersulphone with nominal molecular weights cut off at 9 and 25 kDa had a maximum efficiency to eliminate the microorganism by a log reduction value of 7. Delfini and Formica (2001) reported that the filtration of wines with yeast and bacteria concentration of 3×10^3 cells/ml and 4×10^7 cells/ml using membranes with the pore size 0.22 μm could remove yeast and bacteria up to 100% and 99%, respectively.

2.3.1.3 Membrane morphology

Each membrane will have different numbers of pores per unit area of membrane depending on the membrane morphology, for example, Anopore membrane produced from aluminum oxide will have a high quantity of pores. Ho and Zydney (1999) found that the filtration using membranes with a low quantity of pores resulted in a reduction of permeate flux faster than that with a high quantity of pores due to the faster clogging of the membrane with the lower pore quantity.

2.3.2 Feed characteristics

Effects of feed characteristics are owing to its compositions that are insoluble compounds and soluble compounds. The main factors of insoluble components that affect microfiltration performance are suspended solid size and size distribution, and suspended solid concentration (Persson *et al.*, 1993; Burrell and Reed, 1994; Guell *et al.*, 1999; Czekaj, Lopez, and Guell, 2000; Vyas *et al.*, 2000b; Czekaj Lopez, and Guell, 2001; Eagles and Wakeman, 2002; Metsamuranen *et al.*, 2002).

2.3.2.1 Suspended solid size and size distribution

Solid particles in a sample can affect the clogging of the membrane pore. Burrell and Reed (1994) found that the filtration of beers having suspended solid particle sizes of 0.5-1 μm using a ceramic membrane with the pore size of 0.5 μm had lower permeate flux than the sample having less quantity of suspended solid particle sizes of 0.5-0.6 μm. Since the non-filtrated beer would have a higher quantity of larger particle of 0.5-1 μm, so it could clog the membrane faster and cause the lower permeate flux.

Czekaj *et al.* (2000) studied the filtration of beer samples having 3 molecular sizes; i.e., less than 30 kDa, between 30-80 kDa, and more than 80 kDa, using cellulose acetate membranes, hydrophilic membranes. It was found that the beer sample contained larger molecules had higher resistance.

2.3.2.2 Suspended solid concentration

The filtration of samples having high suspended solids content can cause a reduction of steady state flux.

Vyas *et al.* (2000b) reported that higher concentrations of suspended solids caused higher accumulations of particles at the membrane surface resulting in a higher surface fouling, therefore, caused higher filtration resistances and a lower permeate flux. An increase in surface fouling with suspended solid concentration only occurred at a low suspended solids concentration. Once the suspended solid concentration reached a critical concentration of 2.5 g/l, and then the surface fouling would be constant because an increase in concentration of suspended solid would

interfere the movement of particles to the membrane surface and remove accumulated particles as surface fouling out from the surface. Persson *et al.* (1993) found that the filtration of protein suspension having lactoglobulin's concentrations of 0.1-10 g/l would increase protein adsorptions on the membrane. Guell *et al.* (1999) found that the filtration of samples having suspended yeasts would have lower resistance than that having only proteins because the latter could form internal fouling as a result of protein aggregates. If the size of suspended yeast was higher than that of protein, then it would reduce the internal fouling as the larger size of suspended yeasts could accumulate at the membrane's surface acting as a secondary membrane to help retaining proteins, therefore, the formation of internal fouling would be less. Meanwhile, this effect would cause molecules with the size smaller than membrane pore sizes to pass through more and the permeate flux to increase. Metsamuranen *et al.* (2002) also found that the filtration of yeast suspensions with high suspended solid contents could reduce critical flux.

Compositions of suspended solids can affect the filtration efficiency and filtration characteristics. The filtration of samples having suitable concentrations of suspended solids can increase the permeate flux resulting in an increase in filtration efficiency but a decrease in permeate compositions. Guell *et al.* (1999) found that the filtration of samples having suspended yeasts could affect a reduction of protein compositions of the permeate the first stage of filtration from an increase in protein rejection owing to the high formation of membrane fouling. The filtration of mixed protein yeast suspension having large sizes of suspended yeasts would cause an increase in protein rejection. However, once the concentration of the suspended yeast in the sample is much higher, subsequently it would result in higher resistances and lower permeate flux.

Moreover, it was found that the model turbidity solutions having small sized compositions could affect the membrane fouling as well. Based on the studied of Czekaj *et al.* (2001), it was observed that at the first stage of filtration of model turbid solutions with high turbidity could form internal fouling very fast because of the high concentration of small molecules in the solution.

2.3.2.3 Soluble components

Food has soluble compounds in its compositions such as proteins, polysaccharide, and polyphenol, which are large molecules; therefore, it can affect the filtration efficiency. Eagles and Wakeman (2002) studied the effect of dissolved materials such as starch, α-Glucan and casein in model beer on permeate flux. The model beer consisted of 40 g/l ethanol, 1500 mg/l glycol, 1500 mg/l maltose, 25-100 mg/l citric acid, 50 mg/l catechin, 25-100 mg/l calcium and 50 mg/l ethyl acetate. The 0.2 μm cellulose nitrate membrane was used for this study. The results showed that an increase in concentration of α-Glucan, polymer of glucose, resulted in a significant decrease in permeate flux because the molecules of α-Glucan, which are large molecules could form a gel layer on the membrane's surface as a secondary membrane, and then could induce an increase in the resistance and a decrease in permeate flux. Since α-Glucan molecules were large, therefore they could desorb from the membrane's surface very slow and accumulate to form a gel layer at the membrane surface. Furthermore, it was found that an increase in casein protein in the model beer solution caused a decrease in permeate flux. At that condition, molecules of casein protein had positive charges opposed negative charges of the membrane, so it could create an electrostatic force with the membrane's surface that could assist a

better adsorption at the surface, subsequently increase the formation of membrane fouling and reduce the permeate flux.

2.3.3 Operating conditions

The major factors of operation conditions for the microfiltration are temperature, ΔP, and hydrodynamic above the membrane.

2.3.3.1 Temperatures

An increase in temperature could affect an increase in permeate flux and a decrease in viscosity. Therefore, the diffusivity of the solute would be increase and affect the interaction between the membrane and the solute. However, the temperature could increase up to the limiting temperature. If the temperature is above the limiting temperature, then it could cause a reduction in permeate flux because the high temperature would decrease the solubility of some solutes or cause protein denature. At appropriate temperatures, protein molecules or the solutes could adsorb well at the membrane's surface such as at 30-60 ^{O}C (Cheryan, 1986)

Padilla-Zakour and Mclellan (1993) reported that the filtration of depctinized apple juices using ceramic membranes with the pore size of 0.2 μm at a feed velocity of 9.5 m/s, would have an increase in permeate flux as the temperature increased due to many combined factors of temperatures, transmembrane pressures, and feed velocity, as shown in the following equation:

$$J_m = 283.9 - 0.788\,(1000\Delta P) + 1.64\,(\Delta P \text{ v } T) \qquad (2.22)$$

where J_m = mass permeate flux ($kg/h.m^2$)

T = temperature of feed (^{O}C)

Vladisavljevic *et al.* (2003) studied the filtration of depectinized apple juices using 3 types of inorganic membranes having nominal molecular weight cut off at 30, 50, and 300 kDa and found that an increase in temperature would result in an increase in permeate flux for all 3 types. However, Jiratananon and Chananchai (1996) found that the filtration of passion fruit juices would have a decrease in permeate flux as the temperature increased. The permeate flux had a significant decrease at 40-50 ^{O}C owing to high total resistances caused by the gel formation of pectin in the passion fruit juice on the membrane surface. Besides, pectin could form cross-linking reaction with starch molecules at the membrane surface resulting in an increase in total resistances. Therefore, the nature of feed might give the difference in temperature dependence of permeate flux.

2.3.3.2 Transmembrane pressure (ΔP)

In general, the permeate flux increases as the ΔP increases. At low pressure, the permeate flux governed by the rate at which permeate pass through a porous material. At the higher ΔP, the flux becomes independent of pressure due to concentration polarization and fouling. An increase the ΔP, the flux do not increases (Girard and Fukumoto, 2000). It can be divided to 2 stages (Figure 2.2).

The first stage: permeate control regions, which occur at the beginning of filtration and low pressure. In this stage, permeate flux will be controlled by pressure. An increase in permeate flux will be proportional to an increase in pressure.

The second stage: mass transfer controlled regions, which occur during changes in permeate flux controlled by mass transfer. The permeate flux will not increase in proportion to an increase in pressure. The flux can be described by the film model theory which gives an interpretation of the steady-state mass transfer in the boundary layer.

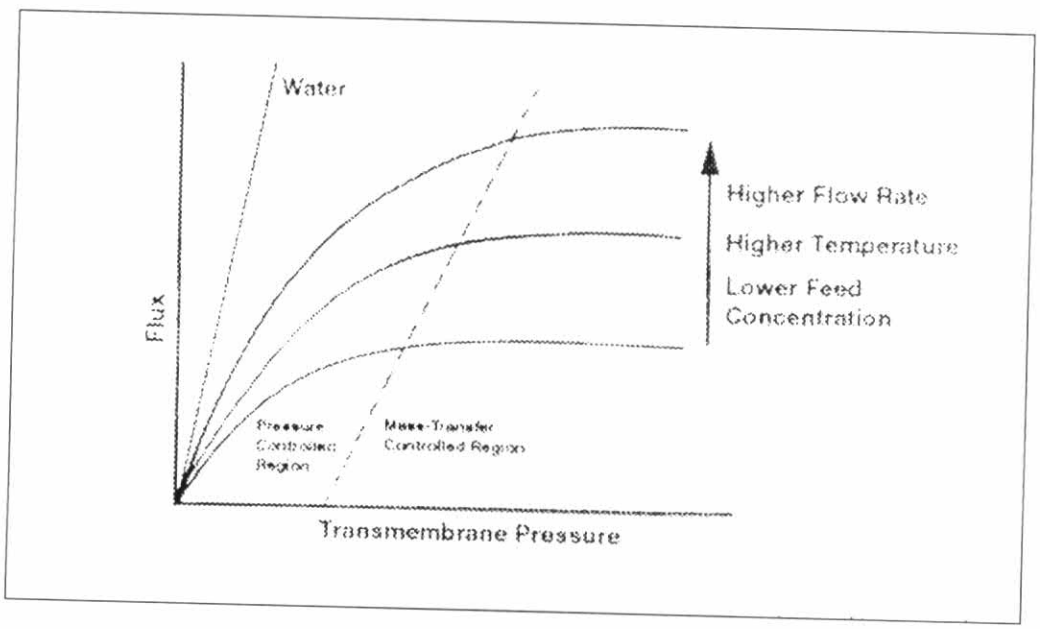

Figure 2.2 Relationship between permeate flux and operating parameters during membrane filtration.

(Girard and Fukumoto, 2000)

Vladisavljevic *et al.* (2003) reported on the filtration of apple juices using inorganic membrane having nominal molecular weight cut off of 30, 50 and 300 kDa that an increase in ΔP resulted in an increase in permeate flux. At a low ΔP, an increase in permeate flux was proportional to an increased pressure due to changes in permeate flux controlled by pressure. When the ΔP increased up to a limiting pressure, an increase in permeate flux was not proportional to the pressure because changes in permeate flux at this stage depended on the mass transfer at the membrane surface, which could affect the formation of concentration polarization and membrane fouling. Changes in permeate flux at this stage was caused by changes in total resistances, a combination of membrane resistance, pore blocking resistance, and cake resistance. Moreover, at high ΔP, the cake could be so compressed that its physical properties could change to be more compacted and less porous resulting in a higher α and a lower permeate flux. This impact would occur on cakes from food and biological samples such as yeasts and bacteria solutions that were compressible cakes (Shimizu *et al.*, 1993).

Increased pressures during the filtration of several fruit juices can have an impact on the permeate flux that it will increase as the pressure increases up to a limiting value, then the increase rate of permeate flux will decrease due to the composition of pectin and several small colloidal particles such as proteins in fruit juices. For proteins having a globular molecular structure, the gel formation at the membrane surface will have loosen structures that allow solutes to pass through, therefore, the permeate flux will increase. On the other hand, for the orange juice, its proteins will have long chain structures of compounds derived from the polymerization of polyphenol and compounds of polyphenol and proteins. The structure can be broken down to be short chains by the pressure and form gel at the

membrane's surface causing a high α. It is also the same impact for a sample having pectin in the composition that large molecules and long chain structures of galacturonic acid can aggregate and form hydrogen bonds. When the pressure increases, the gel layer of pectin solution will be broken down and hydrogen bonds become so compressed that the structure has no void area between chains, thus, the permeate flux will also decrease. Sometimes, the gel layer of pectin can be elastic that the structures can return to their original state of porous structures if an increase in pressure is not high enough to break these structures permanently. According to the study report on the effect of pressures on steady state permeate flux by Vyas *et al.* (2000a), it was found that an increase in transmembrane pressures can affect an increase in steady-state permeate flux up to 100 kPa which is a limiting pressure, and after that the steady-state permeate flux will be constant.

Vernhet *et al.* (2003) reported the effect of pressure on the fouling formation in wines using hydrophilic membranes with the pore size of 0.2 μm. They found that once the pressure for the filtration of crude wine increased (15-100 kPa), then the permeate flux would decrease rapidly in the first stage due to an increase in resistance from membrane fouling. The ratio of fouling resistance to membrane resistance would increase proportional to an increase in pressure because many compositions in the wine could form pore fouling and surface fouling and accumulate as a cake layer on the membrane surface.

2.3.3.3 Hydrodynamic above the membrane

An increase in the feed velocity above the membrane surface results in an increase in the permeate flux. An increase in the feed velocity at the membrane surface will also increase a shearing force acting on particles accumulated on the membrane surface. So the particle accumulation decreases and the mass transfer at the surface increases subsequently, resulting in decreasing in concentration polarization and membrane fouling which lead to increase diffusivity through the membrane. Therefore, permeate flux increases.

De Brujin *et al.* (2002) reported the study of the filtration of apple juices using membranes having nominal molecular weights cut off of 15 and 50 kDa, and found that an increase in sample's velocity could affect an increase in permeate flux. The permeate flux would rapidly decrease at the first stage of filtration due to the adsorption of particles at the membrane's surface causing the boundary layer of the solute and the concentration polarization inducing the mass transfer rate of the solute at the membrane's surface to be increased with a decrease in permeate flux. When the filtration time increased, then the permeate flux would slowly decreased as a result of the formation of membrane fouling, both pore fouling and pore plugging, until the quasi-steady state where the pore blocking occurred and a cake layer was formed at the membrane surface by the particle accumulation. The effect of membrane fouling promoted the formation of the concentration polarization that occurred continuously and caused the permeate flux to decrease constantly as the filtration time increased.

Vladisavljevic *et al.* (2003) studied the filtration of depectinised apple juices using ceramic tubular membranes with a molecular weight cut-off of 300,000, 50,000, and 30,000 Da. The experiments have been carried out over a wide range of ΔP (100-400 kPa), temperatures (20-55 ^{O}C), and feed flow rates (100-900 ml/min). Permeate flux significantly decreased with time until a steady-state was established. The steady-state permeates flux reached a maximum at a ΔP of about 200 kPa. Higher permeate flux was obtained at higher temperatures due to lower permeate viscosity. The steady-state permeate flux was proportional to the feed flow rate raised to powers ranging between 0.22 and 0.31. It also found that log of steady state permeate flux increase in proportional to log (feed flow rate) as the following equation:

$$J_{ss} = k'(Q)^{k''} \qquad (2.23)$$

where J_{ss} = steady state flux (L/h·m^2)

Q = feed flow rate (mL/min)

k' = empirical constant

k'' = empirical constant

The power law parameters k' and k'' were obtained by the nonlinear least-squares regression of the plots between J_{ss} vs feed flow rate.