CHAPTER IV

RESULTS AND DISSCUSSIONS

4.1 Characteristics of fermented liquor, and rice wine

Characteristics of FL and RW filtered with cheesecloth are shown in Table 4.1. The results showed that alcohol content (10% v/v), pH (4.04) and total soluble solids (14 ^{O}Brix) were not significantly different between the FL and RW. Titratable acidity was slightly less for the RW (3.63 g citric acid/100 g) and the FL (3.83 g citric acid/100 g) was not significantly different ($P > 0.05$). The total suspended solids concentration of RW (0.66% w/w) was lower than that of FL (0.79% w/w), presumably as filtering with cheesecloth removed some of the large suspended solids. Microbiological analysis showed that both yeast and bacterial counts were slightly reduced by filtering through cheesecloth. Yeast count was reduced from 3.0×10^8 to 1.2×10^7 CFU/ml, while total bacterial count was reduced from 4.0×10^8 to 3.6×10^7 CFU/ml.

In this research, the fermentation process was not monitored for its completion. However, the alcohol content in fermented liquor was measured after 2 week fermentation and it was higher than 10% v/v, therefore, it could be classified as wine (TISI 208-2544, 2544).

Table 4.1 Characteristics of fermented liquor and rice wine

Characteristics	Fermented liquor	Rice wine
Physicochemical characteristics		
pH[ns]	4.04 ± 0.01	4.04 ± 0.01
Titratable acidity (g citric acid/100 g)[ns]	3.88 ± 0.01	3.63 ± 0.06
Alcohol content (% v/v)[ns]	10.0 ± 0.01	10.0 ± 0.1
Total soluble solid content (°Brix)[ns]	14.0 ± 0.1	14.0 ± 0.0
Total suspended solid content (g/100g)	0.79 ± 0.02[a]	0.66 ± 0.01[b]
Particle size and size distribution of suspended solids (μm)	1-20 and 20-800[a]	1-20 and 20-200[b]
Microbiological characteristics		
Yeast count (CFU/ml)	3.0×10^8 [a]	1.2×10^7 [b]
Total bacteria count (CFU/ml)	4.0×10^8 [a]	3.6×10^7 [b]

Results are the mean values of triplicates measurements and the number following ± are the standard deviations.

[ns] Means in a row are not significantly different ($P \geq 0.05$).

[a, b] Means with same letters in a row are not significantly different ($P \geq 0.05$).

In Figure 4.1, the particle size distributions of FL and RW were bimodal. The FL had particles between approximately 1 and 20 μm, as well as particles between 100 and 1000 μm. The RW also had particles between 1 and 20 μm, but the second peak occurred between 30 and 200 μm. Obviously, initial filtering with cheesecloth drastically reduced the particles above 100 μm. For comparison, gelatinized rice starch granules are typically on the order of 6-16 μm (Jacquier *et al.*, 2006), yeast 2-8 μm (Robinson *et al.*, 2000; Wichramasinghe *et al.*, 2004), and bacteria 0.1-3 μm (Robinson *et al.*, 2000). This would account for much of the particles under the lower particle size peak. In the peak with larger size, one might expect to rice tissue and aggregated cell fragments, as well as any aggregated starch, or cellulose.

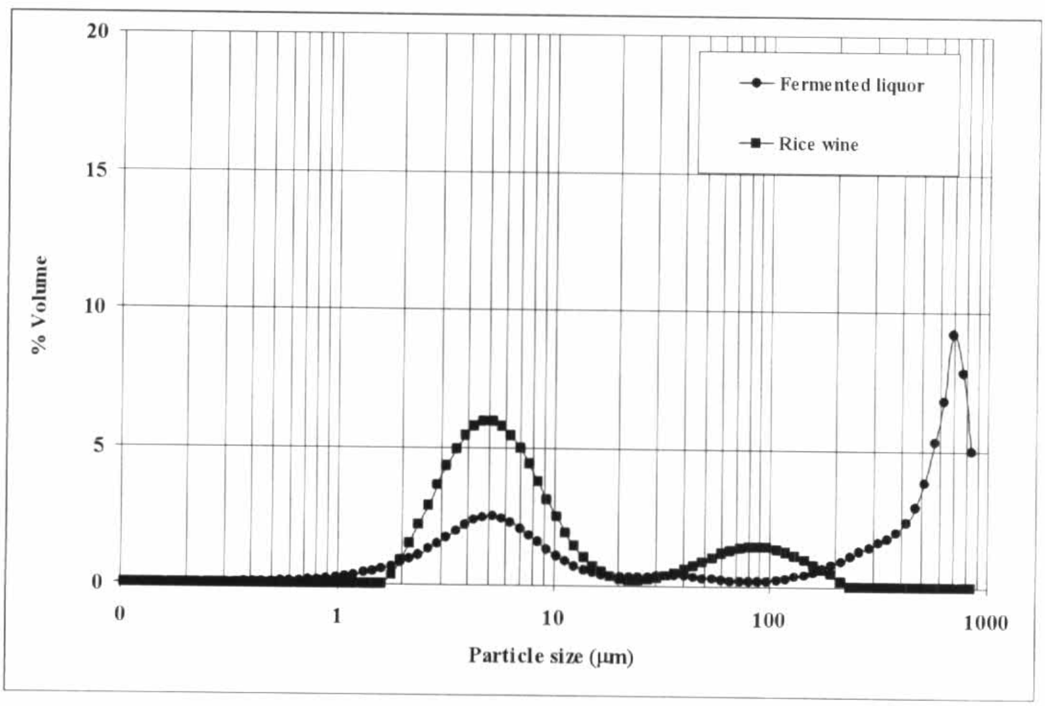


Figure 4.1 Size of suspended solids in fermented liquor and rice wine.

These results show that RW still contains live yeast and bacteria as well as suspended solids (Table 4.1). Thus, secondary fermentation or other microbial activity can cause spoilage, off flavor development or bottle explosion from built-up carbon dioxide gas. In addition, suspended solids from the large particle fraction can continue to settle and form a sediment layer at the bottom of the bottle. The active microorganisms could be destroyed by heating, but with some detriment to the wine flavor. Thus, subsequent sections will deal with the use of microfiltration for cold-sterilization and removal of suspended materials. First, however, we examined the use of pretreatments to decrease the load of microorganisms and other suspended matter prior to microfiltration, with the aim of optimizing the efficiency and life of the microfiltration membranes.

4.2 Effect of solid-liquid separation methods on microfiltration performance, microfiltered rice wine characteristics and taste

Three different solid-liquid separation methods were tested, including filtering the FL through 18 mesh cheesecloth, allowing it to sediment for 3 hours at 20 ^{O}C, or by centrifuging it at 3000xg for 30 min. These approaches were selected as they could be readily implemented in even small-scale wineries. The properties of the RW are shown in Table 4.2 and include alcohol content, titratable acidity, pH, total soluble solids, total suspended solids, particle size distribution (in terms of volume ratio of particles smaller than 20 μm to particles larger than 20 μm, and yeast and bacteria counts.

Table 4.2 Effect of solid-liquid separation methods on rice wine characteristics

Characteristics	Separation methods		
	Filtration through cheesecloth	Sedimentation	Centrifugation
Physicochemical characteristics			
pH[ns]	4.04 ± 0.01	4.04 ± 0.01	4.04 ± 0.01
Titratable acidity (g citric acid/100 g)[ns]	3.64 ± 0.06	3.67 ± 0.06	3.68 ± 0.03
Alcohol content (% v/v)[ns]	10.0 ± 0.1	10.0 ± 0.1	10.0 ± 0.1
Total soluble solid content (°Brix)[ns]	14.0 ± 0.0	14.0 ± 0.0	14.0 ± 0.0
Total suspended solid content (g/100g)	0.66 ± 0.01[a]	0.12 ± 0.01[b]	0.02 ± 0.01[c]
Particle size and size distribution of suspended solids (μm)	0.3–20 and 20–300[a]	0.3–20 and 20–70[b]	0.3–20[c]
Volume ratio of particles smaller than 20 μm to particles larger than 20 μm	80:20[c]	97:3[b]	100:0[a]
Microbiological characteristics			
Yeast count (CFU/ml)	1.2×10^{8}[a]	9.0×10^{6}[b]	5.3×10^{5}[c]
Total bacteria count (CFU/ml)	3.6×10^{7}[a]	3.6×10^{6}[b]	2.5×10^{4}[c]

Results are the mean values of triplicates measurements and the number following ± are the standard deviations.

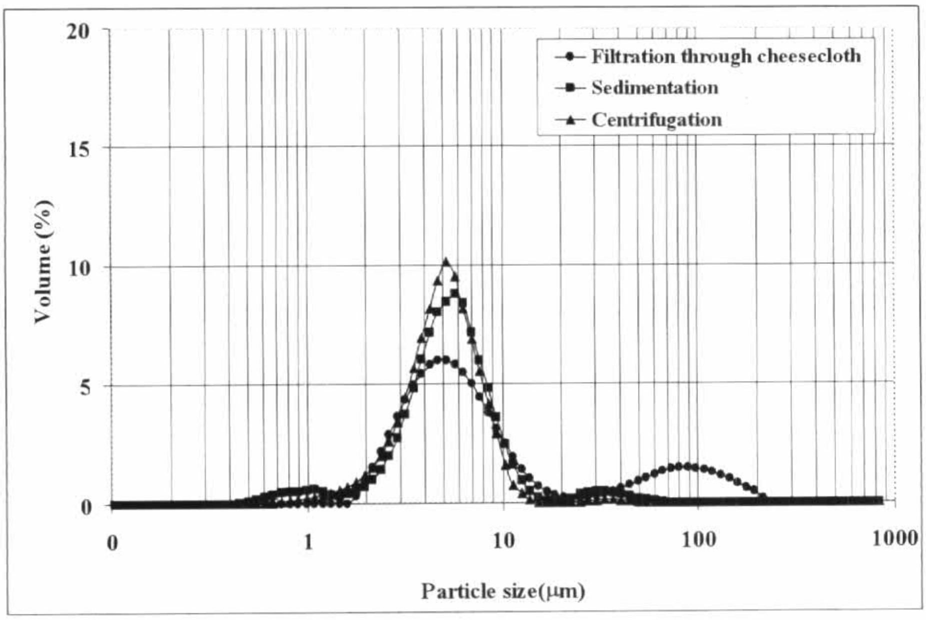
[ns] Means in a row are not significantly different (P ≥ 0.05).

[a, b, c] Means with same letters in a row are not significantly different (P ≥ 0.05).

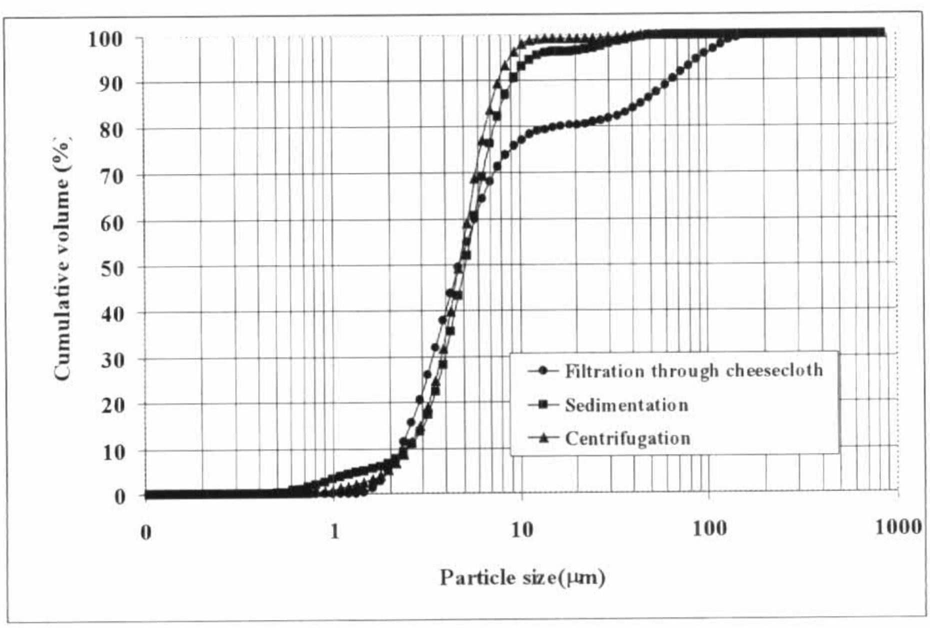
The results showed that alcohol content (10% v/v), titratable acidity (3.64 - 3.68 g citric acid/ 100 g), pH (4.04) and total soluble solids (14 ^{O}Brix) of RW obtained by filtration through cheesecloth (RW_f), sedimentation (RW_s) or centrifugation (RW_c) were not significantly different. Total suspended solids did vary, however, with values of 0.66, 0.12, or 0.02 g/100 g for RW_f, RW_s or RW_c, respectively. Sedimentation or centrifugation were clearly better at reducing suspended solids than cheesecloth alone, with further reductions of 82% or 97% as compared to cheesecloth filtering.

All samples also showed a bimodal size distribution with one common peak (Figure 4.2a, and 4.2b) in the 1-20 μm size range. RW_f had larger particles in the 20-300 μm range, while those for RW_s or RW_c were in the 20-70 μm range. The volume ratio of small (< 20 μm) to large (> 20 μm) particles was 80:20 for RW_f, 97:13 for RW_s, and 100:0 for RW_c. Clearly, sedimentation and particularly centrifugation were more effective at removing large suspended particles.

Microbial counts were also affected by the separation methods. Yeast counts were 1.2×10^8, 9.0×10^6 and 5.3×10^5 for RW subjected to cheesecloth filtration, sedimentation and centrifugation. Bacterial counts were 3.6×10^7, 3.6×10^6 and 2.5×10^4, respectively. Thus, centrifugation was better than sedimentation, which in turn was better than cheesecloth, for removing microorganisms. It is difficult to assess whether this is a direct effect on single, unattached microbial cells, or is due to removal of larger particles to which cells were attached or entrapped. It is clear that preseparation using sedimentation or centrifugation are more efficient at removing large suspended solids and reducing microbial loads than cheesecloth alone.



(a)



(b)

Figure 4.2 Size of suspended solids in rice wine prepared by filtration through cheesecloth, sedimentation, and centrifugation expressed in volume (%) (a) and cumulative volume (%) (b).

To determine the microfiltration performance of RW_f, RW_s, and RW_c, J of each RW was determined from dV/dt and normalized by the effective membrane area. In order to study the effect of separation methods on filtration rate and α (Equation 2.14), J was plotted versus cumulative permeate volume as shown in Figure 4.3. The results showed that the permeate flux during microfiltration for all RW decreased rapidly during initial filtration, and showed more gradually thereafter. The build-up of the filter cake increase resistance to flow, thereby reduces the permeate flux.

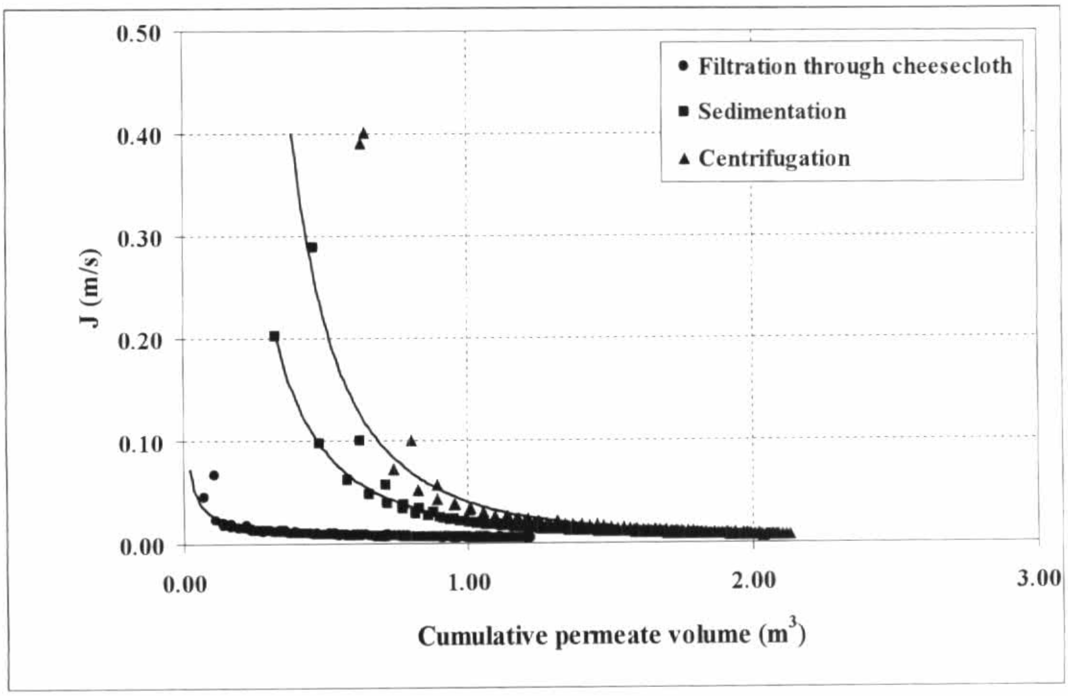


Figure 4.3 Relationship between permeate flux and cumulative permeate volume of rice wines prepared by filtration through cheesecloth, sedimentation, and centrifugation.

Microfiltration condition:

Membrane:	0.45 μm PVDF	
ΔP:	276	kPa
Stirring speed:	100	rpm

In general, filtration was more rapid for samples preseparated with centrifugation and slowest for those treated with cheesecloth only. For example, the initial flux rates (J_o) were 0.078, 0.205, and 0.395 $m^3s^{-1}m^{-2}$ for samples pretreated with cheesecloth, sedimentation or centrifugation, respectively (Table 4.3). This is in keeping with our results on the effect of preseparation on suspended solids and microorganisms. The greater reduction of these materials by centrifugation, and to a lesser extent sedimentation, led to reduced membrane fouling and cake layer formation, and thus less resistance to flow. As noted previously, centrifugation led to much lower total suspended solids, and of this a much greater fraction consisted of smaller size particles. RW_f had the highest suspended solid concentration but the lowest volume ratio of small particles to large particles. Even though, centrifugation was the best method for reduction of both suspended solids size and concentration (Table 4.2). Therefore, gave highest permeate flux (Figure 4.3). However, centrifugation gave the highest initial permeate flux decline (Figure 4.3). The lowest suspended solid concentration of RW_c should not cause the rapid decline of the initial permeate flux. The rapid decline of the initial permeate flux was due to the size of suspended solid, since smaller particles/cells preferentially deposited on the membrane surface which caused flux to be reduced more rapidly (Wickramasinghe *et al.*, 2004; Wakeman and Tarleton, 1999). The separation methods did not affect the equilibrium flux since the permeate flux of the difference in particle size systems were often similar in magnitude (Wakeman and Tarleton, 1999).

Table 4.3 Effect of solid-liquid separation methods on microfiltration performance

Separation methods	Microfiltration performance		
	J_o (m/s)	Slope (s/m^4)	α (m/kg)
Filtration through cheesecloth	$0.078^{c'} \pm 0.015$	$10034^{a'} \pm 169$	$4.33 \times 10^{10\,b'} \pm 0.07 \times 10^{10}$
Sedimentation	$0.205^{b'} \pm 0.006$	$7163^{b'} \pm 225$	$1.70 \times 10^{11\,c'} \pm 0.07 \times 10^{11}$
Centrifugation	$0.395^{a'} \pm 0.005$	$6921^{c'} \pm 156$	$9.87 \times 10^{11\,a'} \pm 0.20 \times 10^{11}$

Results are the mean values of duplicates ± the standard deviations.

$^{a', b', c'}$ Means with same letters in a column are not significantly different ($P \geq 0.05$).

In order to evaluate the effects of separation methods on microfiltration performance, the α was also determined. A plot of 1/J versus cumulated permeate volume is shown in Figure 4.4. The slope of the RW_c was 9.87×10^{11} m/kg, of RW_s was 1.70×10^{11} m/kg, and RW_f was 4.33×10^{10} m/kg. However, centrifugation was removing large suspended solids in RW and minimize fouling prior to microfiltration in RW, but it was not reducing α. The higher α for centrifuged rice wine samples may be attributed to a higher ratio of small to large particles. The suspension contains higher fraction of small suspended solids which leading to greater compact and less porosity cake (Madeni and Fane, 1996; Madeni, 2001).

Presumably, the smaller particles would pack more closely creating a denser cake layer. Therefore, it gave higher resistance to flow per unit mass of the cake layer. However, there was much less total suspended solids in the centrifuged rice wine samples, thus the cake layer thickness was likely much thinner.

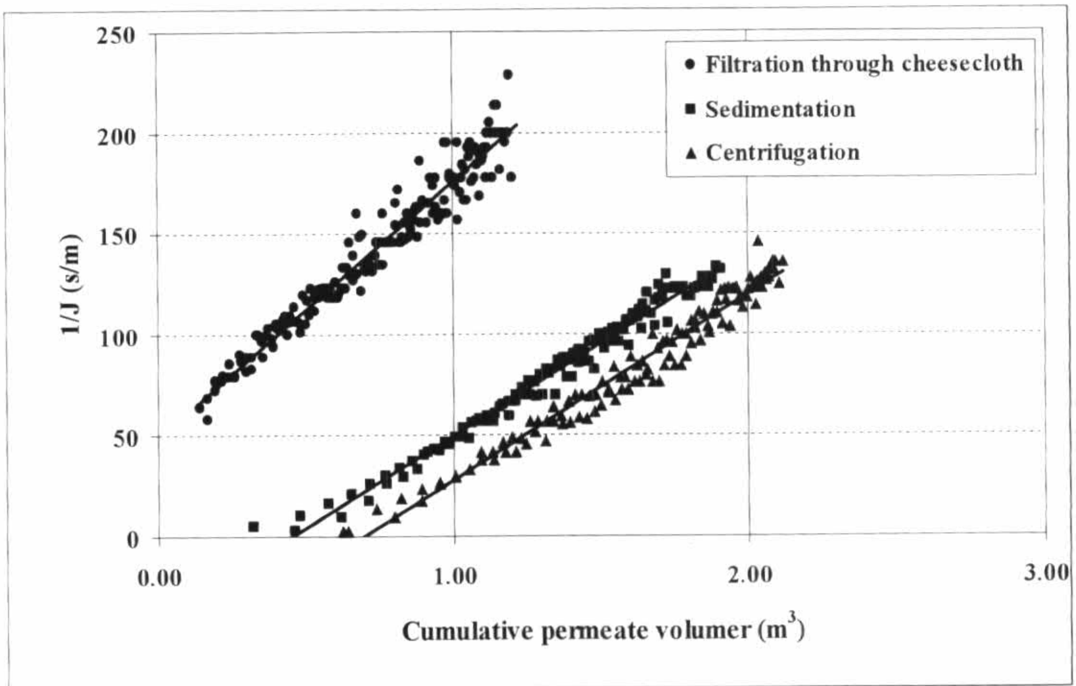


Figure 4.4 Relationship between reciprocal of permeate flux versus cumulative permeate volume of rice wines prepared by filtration through cheesecloth, sedimentation, and centrifugation.

Microfiltration condition:

Membrane:	0.45 µm PVDF	
ΔP:	276	kPa
Stirring speed:	100	rpm

The permeate (MRW) of different preseparated samples were collected from the microfiltration unit and stored in sterile bottles. These were subsequently analyzed for alcohol content, titratable acidity, pH, total soluble solids, total suspended solids, yeast count, bacteria count, and lactic acid bacteria count on the same day (Table 4.4). The results showed that preseparation did not make a significant difference in alcohol content, titratable acidity, pH, or total soluble solids of MRW samples. In addition, for all MRW samples regardless of preseparations, there were no detectable suspended solids, yeast, bacteria, or lactic acid bacteria. This shows that microfiltration was an effective means of clarifying and sterilizing the wine samples. Therefore, it can be used for cold sterilization of RW. In addition, the solid-liquid separation methods did affect the rate of filtration and build-up of a cake layer. However, it did not affect the ability to remove suspended solids or microorganisms.

Table 4.4 Effect of solid-liquid separation methods on microfiltered rice wine characteristics

Characteristics	Separation methods		
	Filtration through cheesecloth	Sedimentation	Centrifugation
Physicochemical characteristics			
pH ^{ns}	4.04 ± 0.01	4.04 ± 0.01	4.04 ± 0.01
Titratable acidity (g citric acid/100 g) ^{ns}	3.64 ± 0.06	3.67 ± 0.06	3.68 ± 0.03
Alcohol content (% v/v) ^{ns}	10.0 ± 0.1	10.0 ± 0.1	10.0 ± 0.1
Total soluble solid content (^{O}Brix) ^{ns}	14.0 ± 0.0	14.0 ± 0.0	14.0 ± 0.0
Total suspended solid content (g/100g) ^{ns}	0	0	0
Microbiological characteristics			
Yeast count (CFU/100 ml)	ND	ND	ND
Total bacteria count (CFU/100 ml)	ND	ND	ND
Lactic acid bacteria count (CFU/100 ml)	ND	ND	ND

Results are the mean values of triplicates measurements and the number following ± are the standard deviations.

ND = Non detectable

^{ns} Means in a row are not significantly different ($P \geq 0.05$).

The Duo-Trio test were carried out to compare the difference in taste of MRW preseparated by filtration through cheesecloth against sedimentation, filtration through cheesecloth against centrifugation, and sedimentation against centrifugation from section 3.5. The twenty experienced panelists evaluated the difference in taste of MRW prepared with different separation methods using the Duo-Trio test. The results were statistically analyzed at significance level of 5% (see Appendix E) as followed O' Mahony (1986) and Meilgaard *et al.* (1987).

It was found that the MRW preseparated by filtration through cheesecloth against MRW preseparated by sedimentation, the MRW preseparated by filtration through cheesecloth against MRW preseparated by centrifugation, and the MRW preseparated by sedimentation against MRW preseparated by centrifugation, were not significant different in taste at significance level of 5% as showed in Table 4.5. Therefore, it was clear that the different separation methods did not affect on taste of MRW as well as and chemical characteristics as shown in Table 4.4.

Table 4.5 Sensory evaluation of microfiltered rice wine using different separation methods prior to microfiltration with 0.45 μm PVDF membrane at 200 kPa.

Test	Difference test between microfiltered rice wine which preseparated by	No of panelists	No of corrected mark	No of false mark	Results*
I	Filtration through cheesecloth and sedimentation	20	7	13	NS
II	Filtration through cheesecloth and centrifugation	20	9	11	NS
III	Sedimentation and centrifugation	20	13	7	NS

NS: Means in a row are not significantly different ($P \geq 0.05$).

* See statistical table in Appendix E.

4.3 Effect of transmembrane pressure, membrane pore size and stirring speed on microfiltration performance and microfiltered rice wine characteristics

To evaluate the effects of operating conditions, three process parameters were selected for study. These included membrane pore size (0.10, 0.22, or 0.45 μm), transmembrane pressure (138, 276, 414, or 552 kPa) and stirring speed (0 or 100 rpm). The RW_s were selected as feed suspension as they had a particle size distribution that covered a typical range for rice wine as shown in Figures 4.1 and 4.2.

Microfiltration studies were conducted at 138, 276, 414, and 552 kPa with membranes of three different pore sizes (0.10, 0.22, or 0.45 μm). The flow rates were determined as described previously, and the α determined from a plot of J versus cumulative permeate volume.

Figure 4.5 shows that α increased with ΔP. According to Equation 2.15, their relationship could be expressed as Equation 4.1 ($r^2 = 0.99$).

$$\alpha = 432261(\Delta P)^{0.98} \qquad (4.1)$$

From Equation 4.1, the cake compressibility of RW cake was 0.98. This indicated that the RW cake was compressible reflected the soft nature of suspended solids derived from the partial hydrolyzed gelatinized rice. The compressibility of microorganism cake was found to be in the range of 0.28 to 1 (Kawakatsu *et. al.*, 1993). The cells of *S. cerevisiae* in rice wine was considered as a compressible particles because their extracellular macromolecules (polysaccharides and lipoproteins) surrounding the cell may become deformed with pressure (Kawakatsu *et. al.*, 1993). The breaking of suspended particles under pressure also caused an increase in α (McCarthy *et al.*, 1998). That is, the cake is more compact and less porous at greater ΔP (Iritani *et al.*, 1991).

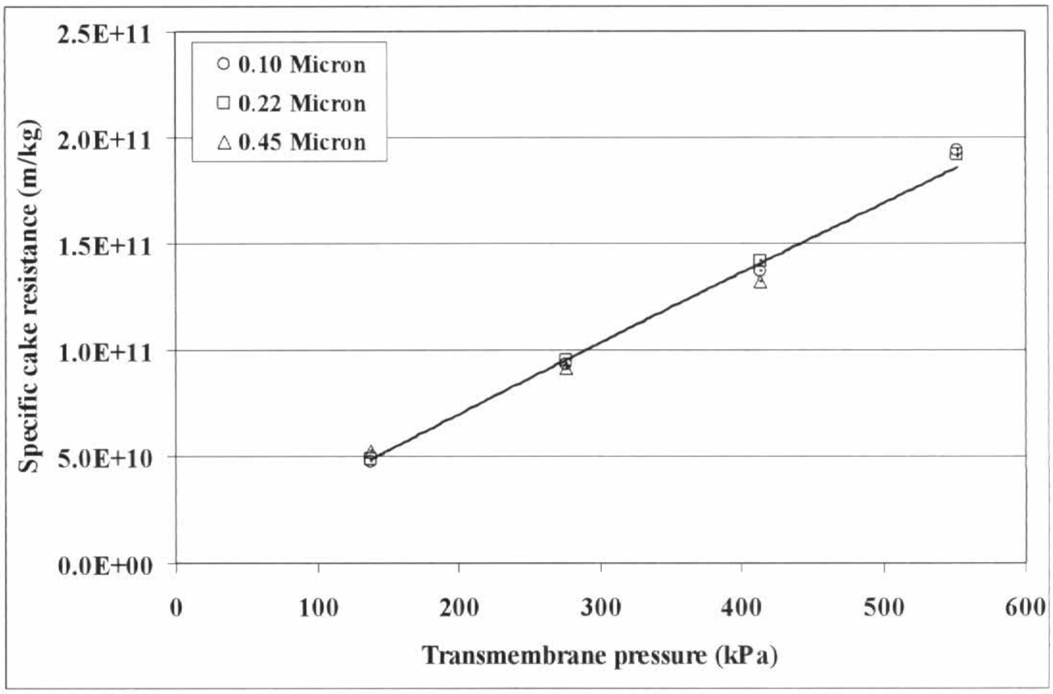


Figure 4.5 Effect of transmembrane pressure on specific cake resistance of rice wine using different membrane pore size.

Figure 4.5 also shows α for samples filtered with 0.10, 0.22, or 0.45 μm PDVF membranes. No significant differences in α were noted due to difference in pore size. The cake layer on the membrane surface acts as a secondary membrane, and these results verify that all suspended solids were trapped and retained in the cake layer regardless of membrane pore size. Therefore, cake layers formed during filtration on different pore-sized membranes do not differ in properties of the cake such as porosity or compact structure.

Table 4.6 shows that the α was not affected by the membrane pore size and stirring speed used in this research. Since, the suspended solids in RW has size larger than the membrane pores, therefore, particles did not clog within the membrane pores. The cake structure and porosity governed the value of α (Lee *et al.*, 2000; Lee *et al.*, 2003; Lodge *et al.*, 2004). Therefore, these results indicated that cake structure and porosity might not be altered by stirring speed and membrane pore size (when membrane pore size was smaller than particle size). The main reason for stirring was to reduce fouling by increasing the turbulence on the feed side of membrane, thus increased permeate flux. However, the permeate flux of MRW did not increase by stirring at 100 rpm. Therefore, stirring speed of 100 rpm did not give enough turbulence flow to reduce fouling and α. Moreover, the filtration time/volume was not long/more enough to see the effect of stirring speed.

Table 4.6 Effect of stirring speed and membrane pore size on specific cake resistance (α) of rice wine.

Membrane pore size (μm)	**ΔP (kPa)**	**Stirring speed (rpm)**	**α (m/kg)**
0.22	138	0	$4.48 \times 10^{10\ a'} \pm 0.18 \times 10^{10}$
		100	$4.85 \times 10^{10\ a'} \pm 0.11 \times 10^{10}$
	552	0	$2.12 \times 10^{11\ b'} \pm 0.09 \times 10^{11}$
		100	$1.91 \times 10^{11\ b'} \pm 0.09 \times 10^{11}$
0.45	138	0	$5.32 \times 10^{10\ a'} \pm 0.23 \times 10^{10}$
		100	$4.90 \times 10^{10\ a'} \pm 0.11 \times 10^{10}$
	552	0	$2.08 \times 10^{11\ b'} \pm 0.11 \times 10^{11}$
		100	$2.00 \times 10^{11\ b'} \pm 0.10 \times 10^{11}$

$^{a', b'}$ Means with same letters in a colume are not significantly different ($P \geq 0.05$).

The effect of operating conditions ($\triangle$P, membrane pore size, and stirring speed) on permeate characteristics are summarized in Table 4.7. As can be seen, alcohol content, pH, and titratable acidity as well as yeast, bacteria and lactic acid bacteria counts, were not significantly different due to processing conditions. Moreover, the results showed there were no detectable suspended solids, yeast, bacteria, or lactic acid bacteria in microfiltered rice wine for any of the operating conditions studied. Thus, it is clear that microfiltration successfully eliminated bacteria, lactic acid bacteria, yeast, and suspended solids, without affecting the basic chemical properties of the RW.

Table 4.7 Effect of transmembrane pressure (ΔP) and membrane pore size on microfiltered rice wine characteristics at stirring speed of 100 rpm.

Operating conditions		Permeate characteristics					
		Physicochemical characteristics			Microbiological characteristics (CFU/100 ml)		
M* (μm)	ΔP (kPa)	pH^{ns}	Titratable acidity^{ns} (g citric acid/100g)	Alcohol content^{ns} (% v/v)	Yeast count	Bacteria count	LAB**
0.10	138	4.02 ± 0.01	3.61 ± 0.02	10.1 ± 0.1	ND	ND	ND
	276	4.02 ± 0.01	3.61 ± 0.04	10.1 ± 0.1	ND	ND	ND
	414	4.02 ± 0.01	3.63 ± 0.05	10.1 ± 0.1	ND	ND	ND
	552	4.02 ± 0.01	3.65 ± 0.06	10.1 ± 0.1	ND	ND	ND
0.22	138	4.02 ± 0.01	3.69 ± 0.04	10.1 ± 0.1	ND	ND	ND
	276	4.02 ± 0.01	3.69 ± 0.02	10.1 ± 0.1	ND	ND	ND
	414	4.02 ± 0.01	3.69 ± 0.02	10.1 ± 0.1	ND	ND	ND
	552	4.02 ± 0.01	3.65 ± 0.03	10.1 ± 0.1	ND	ND	ND
0.45	138	4.02 ± 0.01	3.59 ± 0.02	10.1 ± 0.1	ND	ND	ND
	276	4.02 ± 0.01	3.58 ± 0.03	10.1 ± 0.1	ND	ND	ND
	414	4.02 ± 0.01	3.67 ± 0.05	10.1 ± 0.1	ND	ND	ND
	552	4.02 ± 0.01	3.61 ± 0.07	10.1 ± 0.1	ND	ND	ND

Results are the mean values of triplicates measurements and the number following ± are the standard deviations.

* Membrane pore size

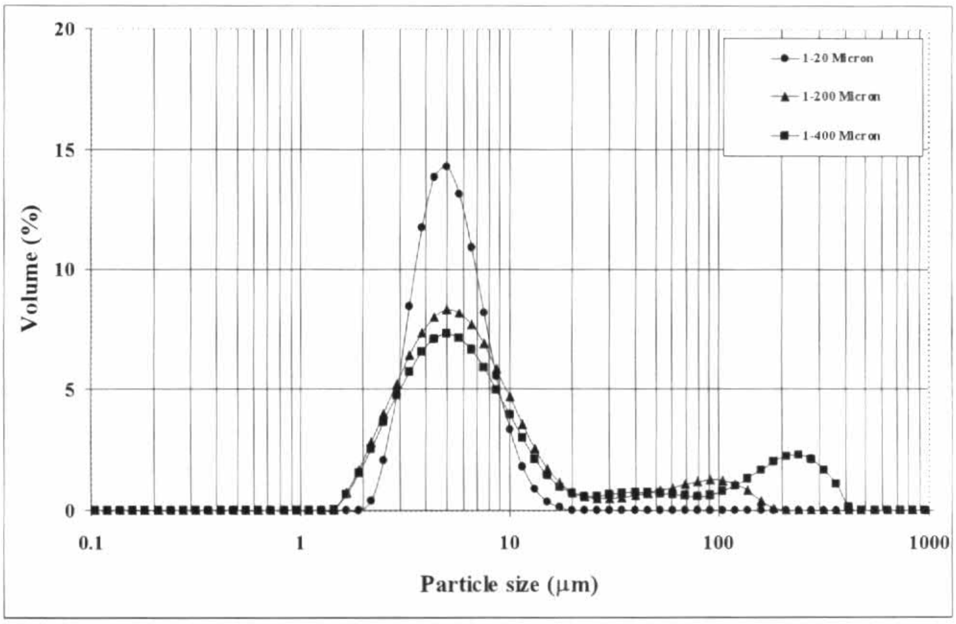
** Lactic acid bacteria count

ND = Non detectable

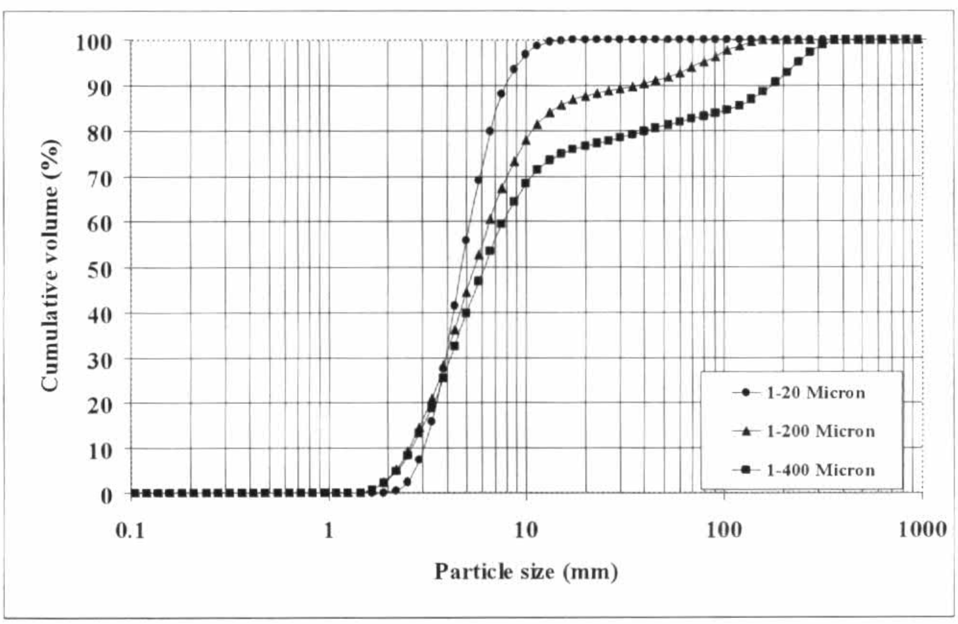
^{ns} Means in a row are not significantly different ($P \geq 0.05$).

4.4 Effect of suspended solid size, size distribution and concentration on specific cake resistance

RW having suspended solids of three different particle size range, which were 1-20, 1-200, and 1-400 μm (Figure 4.6a), were used for this study. While the 1-20 μm sample had a single peak, the 1-200 and 1-400 μm samples showed bimodal size distributions with the most of particles (88% by volume for 1-200 μm sample and 77% for 1-400 μm sample) having diameter in the range of 1-20 μm (Figure 4.6b). For the 1-400 μm samples, about 20% of particles had diameter ranging from 20 to 200 μm and only 10% of particles had diameter ranging from 200-400 μm (Figure 4.6b). The volume weighted mean diameters of particles were 5, 14, and 59 μm, and the mass median diameters were 4.8, 5.5, and 6.6 μm for the 1-20, 1-200 and 1-400 μm samples, respectively.



(a)



(b)

Figure 4.6 Size of suspended solids in rice wine having suspended solid size in the range of 1-20, 1-200, and 1-400 μm expressed in volume (%) (a) and cumulative volume (%) (b).

The α of these samples of various suspended solid concentration in the range of 0.05 to 1.00 wt% were determined (Figure 4.7). The results showed that at the same suspended solid concentration the α of the 1-20 μm sample was the highest and that of the 1-400 μm sample was the lowest. At 1 wt% suspended solid concentration, the α of the 1-20 and 1-200 μm samples were 18.3 and 8.5 times, respectively which higher than that of the 1-400 μm sample. In this range of suspended solid concentration, while the 1-20 and 1-200 μm samples showed an increase in α with suspended solid concentration (r^2= 1.00 and 0.37 , respectively).

The general relationship between suspended solid concentration and α can be expressed as follows:

$$\alpha = j + k\,C \tag{4.2}$$

where C = suspended solid concentration (wt%).

j = empirical constant.

= 3.74×10^{11} for 1-20 μm

= 3.52×10^{11} for 1-200 μm

= 5.75×10^{11} for 1-400 μm

k = empirical constant

= 6.34×10^{11} for 1-20 μm

= 1.63×10^{11} for 1-200 μm

= 0 for 1-400 μm

The suspended solid concentration did not affect the α for the 1-400 μm samples (as k = 0), therefore, the value of j was the mean value of α.

The presence of 20 % of particles with diameter 20-200 μm in the 1-200 μm sample caused a decrease in both α and the dependence of α on suspended solid concentration. The presence of only 10 % of particles having diameter larger than 200 μm (in the 1-400 sample) gave the major decreasing in α and solely contributed to the suspended solid concentration independent phenomena of α. These results indicated that presence of at least 10 % of the particles with diameter higher than 200 μm was required for high microfiltration performance as their acted as self filtering aids. This can be explained by the fact that the presence of the particles having diameter larger than 20 μm in rice wine could promote a decrease in α resulting in an increase of permeate flux. It was found that the flocs of 40 μm gave lower α than the flocs of 10 μm, since the smaller floc formed more compact cake (Tanaka *et al.*, 2001; Lee *et al.*, 2003; Lee *et al.*, 2005).

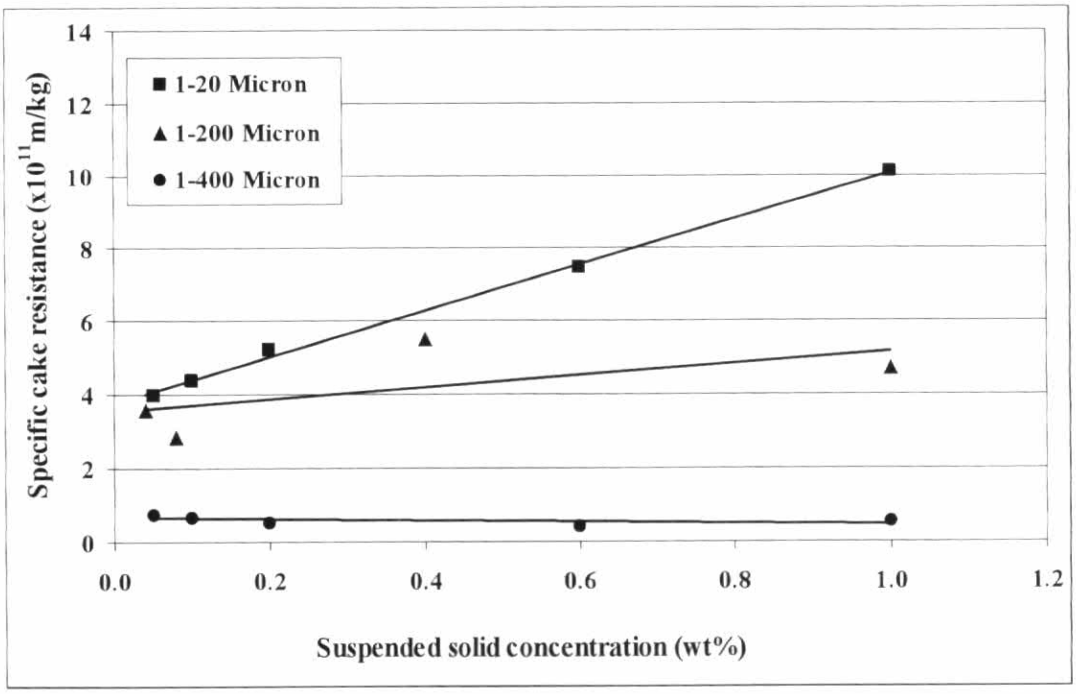
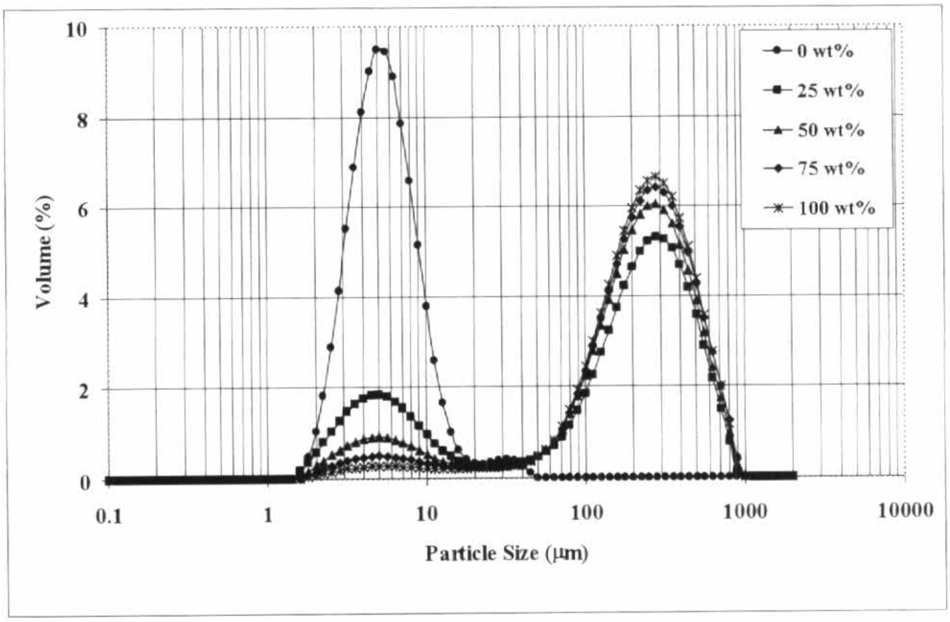


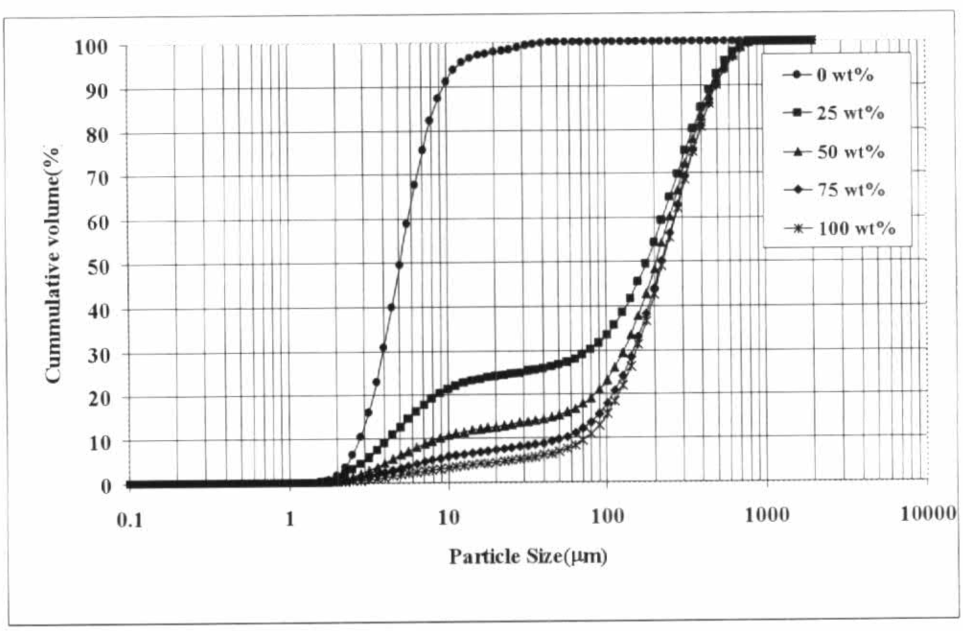
Figure 4.7 Effect of suspended solid concentration on specific cake resistance of rice wine having suspended solid size in the range of 1-20, 1-200, and 1-400 μm.

The result suggested that suspended solids of size larger than 20 μm could be used as self filtration aid to reduce the α. Figure 4.6a shows that the second peak of 1-200 μm and 1-400 μm started at particle size about 45 μm. Therefore, suspended solids of size larger than 45 μm could be added or resuspended into rice wine prior to microfiltration as self filtration aid. Therefore, the effect of concentration of "L" fraction (see section 3.2.3 page 33) in terms of gram of suspended solids having size larger than 45 μm per 100 gram of total suspended solids in rice wine (W) on α was determined.

The size and size distribution of suspended solids in five RW samples having different "L" fraction concentration (W) were determined (Figure 4.8). The volume weighted mean diameters of particles were 6, 206, 243, 255, and 260 μm, while the mass median diameters were 5, 180, 206, 223, and 228 μm for W of 0, 25, 50, 75, and 100 wt%, respectively. For all total suspended solid concentration (0.2, 0.5 and 1.0 wt %), the α decreased as W increased (Figure 4.9).



(a)



(b)

Figure 4.8 Size of suspended solid in rice wine having gram of suspended solid having size larger than 45 μm per 100 gram of total suspended solids of 0, 25, 50, 75, and 100 wt% expressed in volume (%) (a) and cumulative volume (%) (b).

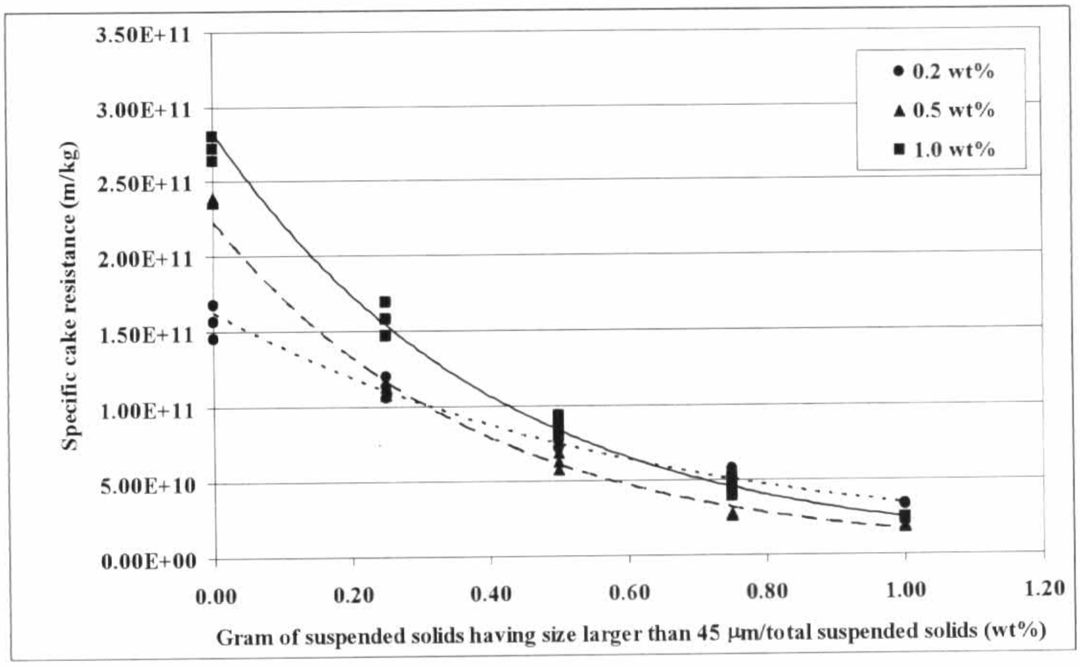


Figure 4.9 Effect of gram of suspended solids having size larger than 45 μm per 100 gram of total suspended solids on specific cake resistance for suspended solid concentration of 0.2, 0.5, and 1.0 wt%.

The general relationship between α and W could be explained using a empirical model ($r^2 > 0.99$) and could be expressed in simplify form as follows:

$$\alpha \quad = \quad x\,(1/e^{yW}) \qquad (4.3)$$

where W = gram of suspended solids having size larger than 45 μm per 100 gram of total suspended solids in rice wine

x = empirical constants

= 1.62×10^{11} for suspended solid concentration 0.2 wt%.

= 2.22×10^{11} for suspended solid concentration 0.5 wt%.

= 2.83×10^{11} for suspended solid concentration 1.0 wt%

y = empirical constants

= 1.55 for suspended solid concentration 0.2 wt%.

= 2.59 for suspended solid concentration 0.5 wt%.

= 2.44 for suspended solid concentration 1.0 wt%

These empirical models can be used to predict the α of RW samples having suspended solid concentration and size distribution in the range between 0.2 to 1.0 wt% and 1.0 to 1000 μm, respectively. In order to increase microfiltration performance, this information has a potential application for minimizing α in microfiltration of rice wine by mixing large suspended solids. Even though, preseparation of large suspended solids prior to microfiltration in rice wine samples can minimize fouling in feed suspension, these did not reducing α. Since, these RW contained low fraction of large suspended solids which formed high compact and less porosity cake. Therefore, an increase in large suspended solids fraction in rice wine samples prior to microfiltration, can be used for maximize microfiltraion performance.

Figure 4.9 also shows that α increased with total suspended solid concentration at 0 wt% of W, and then effect of total suspended solid concentration on α started to diminish when $W \geq 50$ wt%. The Carman-Kezeny equation (Equation 2.17) states that the cake resistance is a function of porosity of the cake layer (Fane, 1984; Foley, 2006). The smaller particles formed more compact (less porosity cake) (Tanaka *et al.*, 2001; Lee *et al.*, 2003, Lee *et al.*, 2005). The α was found to be a reciprocal function of the square of geometric mean diameter of particles as shown in Equation 2.22 (Endo and Alonso, 2001).

To illustrate to effect of suspended solid size, size distribution and concentration on α, numbers of suspended solid particles per volume of rice wine (N), volume of rice wine per suspended solid particle (v_{RW}) and cross sectional area per suspended solid particle (A_p) were calculated. At the same total suspended solid concentration (1 wt%) the weight by volume concentration can be calculated from

$$C_{w/v} = C\rho_{RW} \tag{4.4}$$

when $C_{w/v}$ = weight by volume concentration of total suspended solids (g_{TSS}/100 ml RW)

C = weight by volume concentration of total suspended solids (g_{TSS}/100 g RW)

ρ_{RW} = density of rice wine (g/ml)

TSS = total suspended solid

Assuming spherical particles of constant density, the volume of a suspendened solid particle and the numbers of particles per 100 ml of rice wine can be calculated for different particle size.

$$v_{TSS} = (4/3)\pi(D/2)^2 \qquad (4.5)$$

when v_{TSS} = volume of a suspendened solid (ml)

D = Diameter of a suspended solid (cm)

The numbers of particles per 100 ml RW can be calculated as follow as:

$$\begin{aligned} N &= (C_{w/v}/\rho_{TSS})/\ v_{TSS} \qquad (4.6) \\ &= (C_{w/v}/\rho_{TSS})/(4/3)\pi(D/2)^3 \\ &= (C\rho_{RW}/\rho_{TSS})/(4/3)\pi(D/2)^3 \end{aligned}$$

when N = numbers of particles per volume of rice wine

ρ_{TSS} = density of total suspended solid (g/ml)

At a constant total solid concentration, $(C\rho_{RW}/\rho_{TSS})/(4/3)\pi(1/2)^3$ is constant, then the ratio of N for different particle having size of D_1 and D_2 can be calculated

$$N_1/N_2 = D_2^3/\ D_1^3 \qquad (4.7)$$

when N_1 = numbers of particles per 100 ml of rice wine for particle having size of D_1

N_2 = numbers of particles per 100 ml of rice wine for particle having size of D_2

Table 4.8 shows relative numbers of particles per volume of rice wine (N_R), relative cross sectional area per particle (A_{PR}), relative volume of rice wine per particle (v_{RWR}), and relative total surface area on each layer of membrane surface occupied by particles for different suspended solid size and concentration calculated for $D_2 = 3D_1$. From Table 4.8, at the same total suspended solid concentration, rice wine contained smaller particle size had more numbers of particles (N_R) which resulted in the higher total occupied surface area on each layer of membrane surface (A_{TR}). Therefore, it left less void area on membrane which caused higher in the α. When total suspended solid concentration increased, both N_R and A_{TR} increased resulting in an increasing of the α. These number were used to draw the pictures (Figures 4.11 and 4.12), which depict the effect of suspended solid size (Figure 4.10), size distribution (Figure 4.11) and total solid concentration (Figure 4.12) on void area/size of the cake on the surface of membrane which affects the α. In Figures 4.13 and 4.14, pictures of hexagonal pack of spherical particles were drawn to give clear picture of the effect of particle size on void size. However, random pack was more likely to occur in real system. Void size of hexagonal pack is smallest among various pack while that of random pack is largest. The average diameter of the largest sphere which can pass through void of hexagonal pack (Figure 4.14) or average void size (D_{void}) for sample having different W was estimated from their volume weighted mean diameters. D_{void} was 0.96, 33, 39, 41, and 42 μm for W of 0, 25, 50, 75, and 100 wt%, respectively. Since D_{void} of W ≥ 50 wt% was about 86 times larger than membrane pore size (0.45 μm), therefore α was small and was not affected by total suspended solid concentration.

Table 4.8 Number of particles per volume of rice wine, cross sectional area per particle and volume of rice wine per particle for different suspended solid size and concentration

Sample number (i)	Diameter of suspended solid	Suspended solid concentration	N_R	v_{RWR}	A_{pR}	A_{TR}
1	$D_1 = 1$	1	27	1.0	1	1
2	$D_2 = 3$	1	1	26.0	9	0.33
3	$D_1 = 1$	2	54	0.5	1	2
4	$D_2 = 3$	2	2	13.0	9	0.66

N_R = relative numbers of particles per volume of rice wine = $N_i/N_{i=1}$

N = numbers of particles per volume of rice wine

A_{pR} = relative cross sectional area per particle = $A_{pi}/A_{pi=1}$

A_p = cross sectional area per particle

A_{TR} = relative total surface area on each layer of membrane surface occupied by particles = $A_{Ti}/A_{Ti=1}$

A_T = total surface area on each layer of membrane surface occupied by particles

v_{RWR} = relative volume of rice wine per particle = $v_{RW i} / v_{RW i=1}$

v_{RW} = volume of rice wine per particle of sample number i

Subscript:

i = property of sample number i

Numbers shown in this are roundup number.

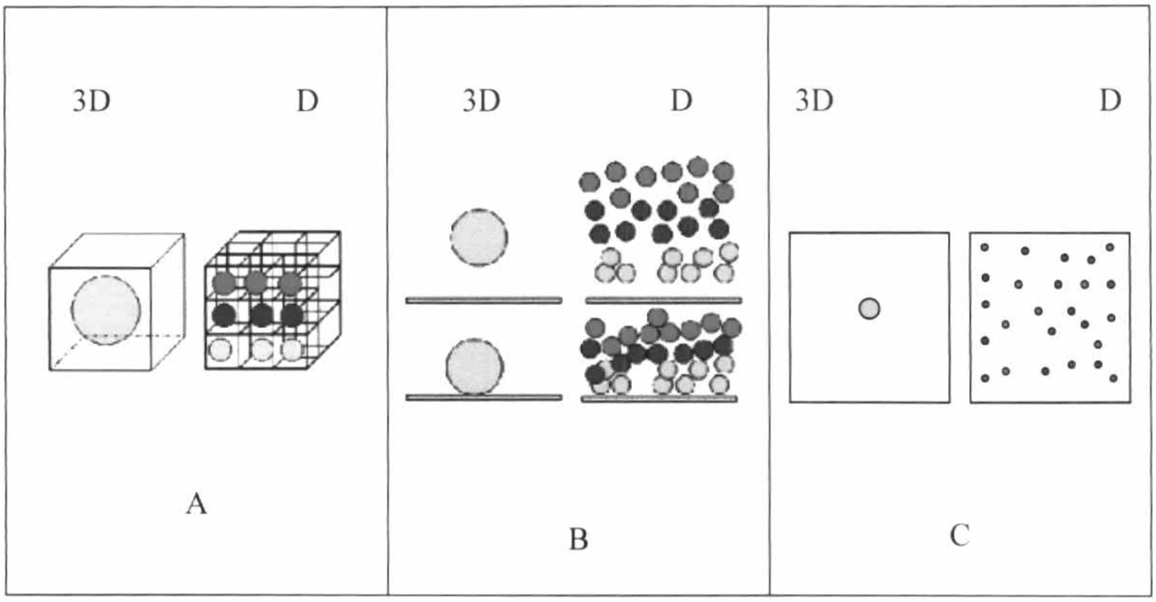


Figure 4.10 Effect of suspended solid size (D and 3D) on void size of the cake on the surface of membrane for the same total solid concentration, feed volume and membrane surface area.

A Shows number of particles for the same total solid concentration and feed volume.

B Shows flowing down of particles to deposit on the membrane surface.

C Shows depositing of particles on membrane surface.

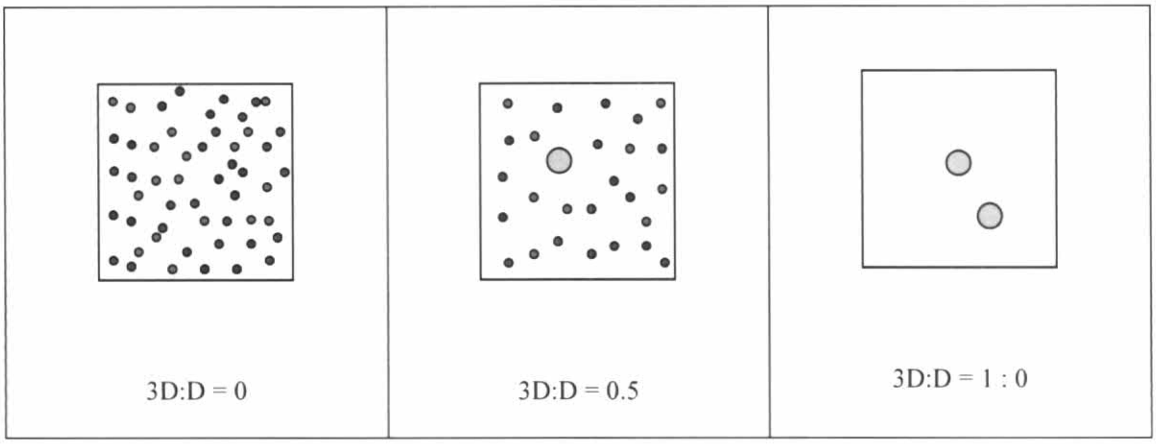


Figure 4.11 Effect of suspended solid size (D and 3D) and size distribution on void size of the cake on the surface of membrane for the same total solid concentration, feed volume and membrane surface area.

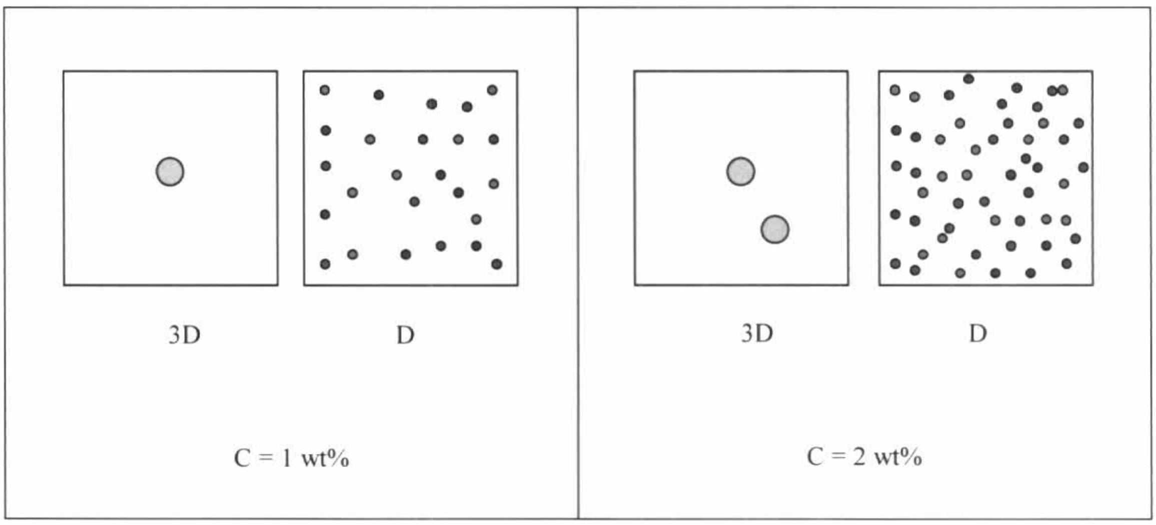


Figure 4.12 Effect of suspended solid size (D and 3D) and total suspension concentration (C) on void size of the cake on the surface of membrane for the same feed volume and membrane surface area.

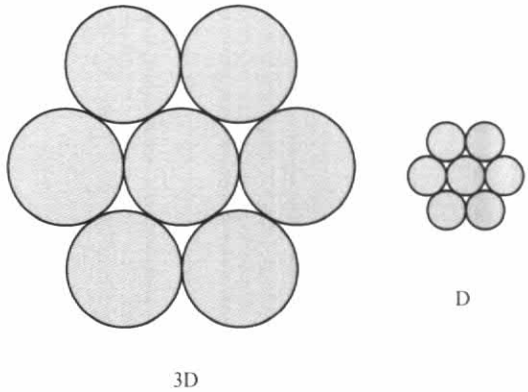


Figure 4.13 Effect of suspended solid size (D and 3D) on void size for hexagonal pack.

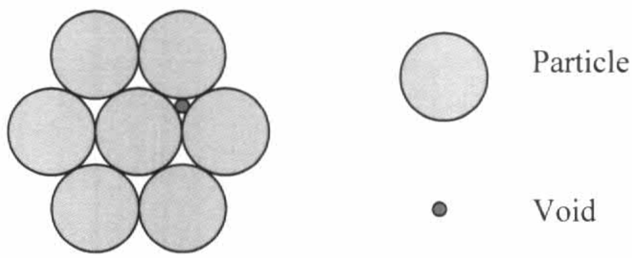


Figure 4.14 Suspended solid size and void size for hexagonal pack.