

Short communication

Limiting permeate flux in the clarification of untreated starfruit juice by membrane ultrafiltration

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Abstract

This paper describes the limiting flux of untreated starfruit juice obtained during clarification using membrane ultrafiltration. Experiments were conducted in a stirred cell unit using membranes of 25,000 MWCO at 30°C for a range of juice concentration varying from 0.46 to 6.5 wt.%. In addition to the limiting flux behaviour, the relationship between the concentration of solute in the bulk solution and the gel layer was also established. The measured experimental data was analysed using the gel model. In general, it was concluded that the gel model could not satisfactorily describe the clarification of starfruit juice. The observed deviation from the model was mainly due to the high pectin content of the juice. © 1998 Elsevier Science S.A. All rights reserved.

Keywords: Permeate flux; Untreated starfruit juice; Membrane ultrafiltration

1. Introduction

Starfruit (*Averrhoa carambola* L.), as it is sometimes called, is an exotic tropical fruit that is grown on a large scale in Malaysia. The fruit has a high nutritional value and in many cases, it is consumed in its fresh form. In order to improve its marketability, especially for the export market, efforts have been made to produce value added products from the fruit. Among the potential products which are expected to receive favourable response among the consumers is clarified juice.

A viable technique which can be used to clarify starfruit juice is by membrane separation where the process is normally conducted at low temperature. This is extremely important as fruit juice processing is sensitive to elevated temperatures. In addition, membrane processing requires low energy input, thus making it an attractive choice for this purpose. Membrane separation has been applied successfully in the clarification of many fruit juices including that of apple [1], pear [2] and kiwi [3] as well as in the concentration of nutritious vegetable [4]. Based on this successful practice, it is evident that the application of membrane process to clarify starfruit and other tropical fruit juices seems promising but has not been seriously explored. As a first step to evaluate its commercial viability, this work was undertaken to study the

limiting flux behaviour of untreated starfruit juice when subjected to ultrafiltration. A greater emphasis was placed on the role of pectin, which is a major constituent of untreated fruit juice. The suitability of the ideal gel polarisation model to describe the observed data was also assessed.

1.1. Gel polarisation model

As part of an initial effort to understand the process, the simple gel polarisation model was used to analyse the observed data. Details of the model are available elsewhere [5] and will not be discussed here. Only the salient points of the model will be highlighted. In general, the model postulates the formation of a layer of gel on the surface of the membrane that would increase its overall resistance. Gel formation is a significant phenomenon in ultrafiltration due to high flux through the membrane, high retention and low diffusivity of the macromolecules. According to the model, the gel concentration, C_g , is independent of the bulk concentration, C_b . The model is also capable of describing the occurrence of limiting flux. In this case, the permeate flux would increase to a limiting value as the transmembrane pressure, TMP, across the membrane is increased. It has been shown that the value of the limiting flux, J_∞ , is given by:

$$J_\infty = k \ln \left(\frac{C_g}{C_b} \right) \quad (1)$$

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where k is the mass transfer coefficient. From this relationship, a plot of J_w against $\log(C_b)$ would yield a straight line of slope $(-k)$. Eq. (1) is valid if the solute concentration in the permeate is zero.

2. Experimental

2.1. Experimental set-up

A bench scale stirred membrane cell was used to determine the limiting flux profiles during clarification of untreated starfruit juice by ultrafiltration. Details of the experimental set up are shown in Fig. 1. The cylindrical cell which was constructed from perspex was designed to provide an effective filtration area of 15 cm². It was fitted with a pressure relief valve and a nitrogen line which was connected to a high pressure nitrogen tank. The entire cell was placed inside a constant temperature bath. With this arrangement, it was possible to vary the cell temperature and pressure. However the results discussed in this paper were those obtained at 30°C. Flat polysulfone membranes of 25,000 MWCO were used. These membranes were obtained from Millipore, USA. For a typical run, about 400 ml of freshly prepared juice was charged into the cell. The system was then allowed to stabilise at 30°C for about 10 min before the pressure was quickly raised and maintained at the desired value to indicate the beginning of the experiment. The permeate flux was determined by the volume collected within a specified time. To minimise the variations in feed concentration during the course of the experiment, additional fresh juice sample was added at intervals when its original volume in the cell had reduced by about 10 vol.%. At the end of the experiment, the membrane was removed for the determination of the gel layer concentration. A new piece of membrane was used to start an experiment for a new set of operating conditions.

2.2. Preparation of raw material

Selected fresh starfruits were washed and sliced into cubes and homogenised using a household blender. The resulting mush was pressed out using a muslin cloth. The juice was

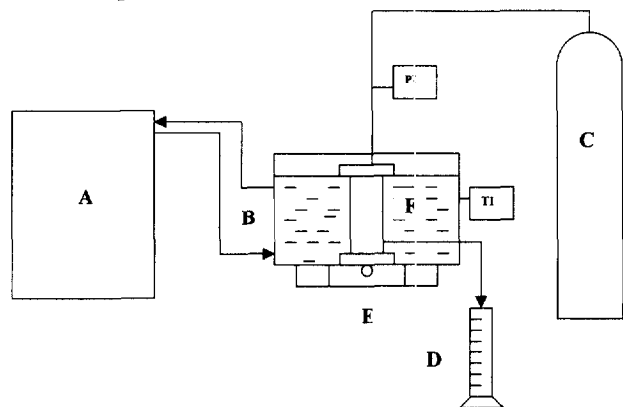


Fig. 1. Schematic diagram of the experimental set-up.

further passed through a 500- μ m sieve to separate and remove any remaining fibre in the sample. Prepared juices were kept frozen at -20°C in a deep freezer. The samples were quickly thawed under running tap water before each experiment. Analysis of the juice is given in Table 1.

2.3. Determination of gel layer and feed concentration of juice

The gel layer concentration was determined at the end of each experiment. The spent membrane, together with the gel, was dried in an oven at 60°C for about 48 h until no further weight change was observed. From the difference in the weight before and after drying, minus the weight of the membrane, the gel concentration was calculated. A similar procedure was performed to calculate the feed concentration by drying a known weight of fresh juice in a crucible.

3. Results and discussion

Before each piece of membrane was used in the experiment, it was first characterised based on the observed water flux as a function of TMP. In general, all membranes gave identical water flux profiles which increased linearly with increasing TMP. From the data, it was estimated that the hydraulic resistance of the membrane, R_m was 0.323 (h m² kPa/l). The variation in the permeate flux with TMP is shown in Fig. 2. For the different feed concentrations investigated, which were, in this case, assumed to be equal to the bulk concentration C_b , the maximum fluxes appeared to occur at TMP = 154 kPa and were independent of the feed concentration. Contrary to the behaviour of protein solutions reported by a number of investigators [6,7], constant values of limiting flux were not observed in this case. Instead, a bell-shaped permeate flow characteristic similar to that obtained from the ultrafiltration of pear juice reported by Kirk et al. [2] was obtained. These results suggested that the nature of the polarised gel layer is influenced by the feed material. Proteins are spherical or globular molecules. The spaces within a protein gel layer are never completely closed, thus allowing the permeate to pass through to give a flux plateau above the optimal TMP. On the other hand, fruit juices are normally rich in pectin. Pectin is a chain-like molecule aggregate of repeating units of galactouranic acid bonded by bridges of hydrogen

Table 1
Typical analysis of starfruit juice

Component	wt. %
Water	93.7
Protein	0.71
Fat	0.10
Carbohydrates	4.94
Pectin	0.15
Ash	0.40

[8]. It has been reported [2] that the pectin gel layer is elastic in nature as evident by the observed partial flux restoration through relaxation of pressure. They suggested that any partial collapse or compression of the gel layer would lead to closure of spaces in the 'net-like' matrix structure of the pectin layer. At pressures exceeding the optimum value, the structure collapsed completely, thus effectively sealing the spaces which would eventually result in flux deterioration in a bell-shaped profile, as shown in Fig. 2. This bell-shaped permeate flux is related to the shape and character of the constituent material which, in turn, affect the molecular orientation and arrangement within the layer. Thus, it is obvious that deviation from the idealised gel model has been observed in this case.

To further confirm that the idealised model could not fully describe the process, the variation in the limiting permeate flux is plotted against the bulk concentration, C_b , (on log scale) according to Eq. (1). The plot is shown in Fig. 3. From Eq. (1), a linear relationship would be observed between the limiting flux and $\log C_b$, provided the mass transfer coefficient, k , is a constant. The value of k depends on the thickness of the boundary layer, δ , and diffusion coefficient, D . In this case, δ remained constant throughout the study since the movement of fluid was only through the membrane and not along its surface that would have affected the boundary layer thickness. The value of D can be taken as a constant since the experiment was conducted under isothermal condition. In addition, the effect of concentration on D is minimal since only dilute solutions of fruit juice were

employed (0.46–6.5 wt.%). As indicated earlier, Eq. (1) is only valid if the solute concentration in the permeate is zero. From the analysis made on the amount of solute in the permeate, it was found that its concentration is less than 8% of that in the feed. In this respect, and without introducing any

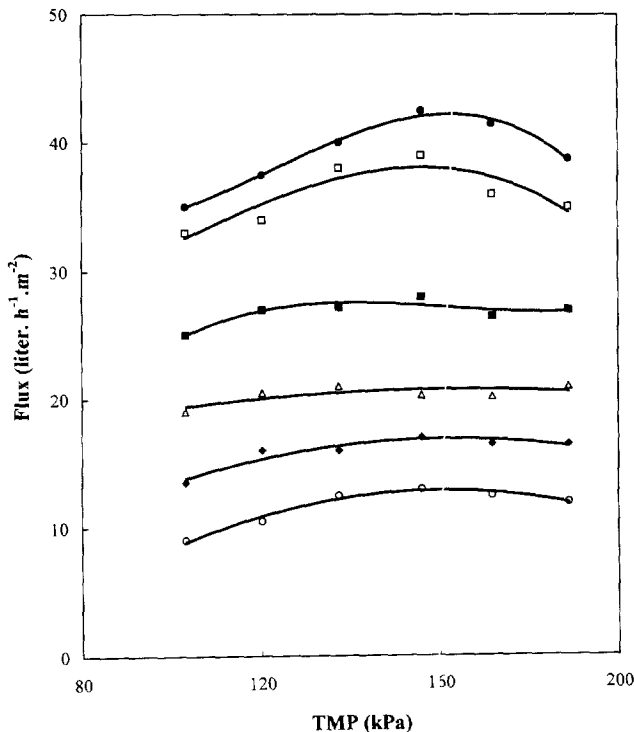


Fig. 2. Variation of permeate flux with pressure at different initial bulk concentrations of 0.46% wt (●), 0.54% wt (□), 1.42% wt (■), 2.04% wt (△), 3.45% wt (◆) and 6.56% wt (○).

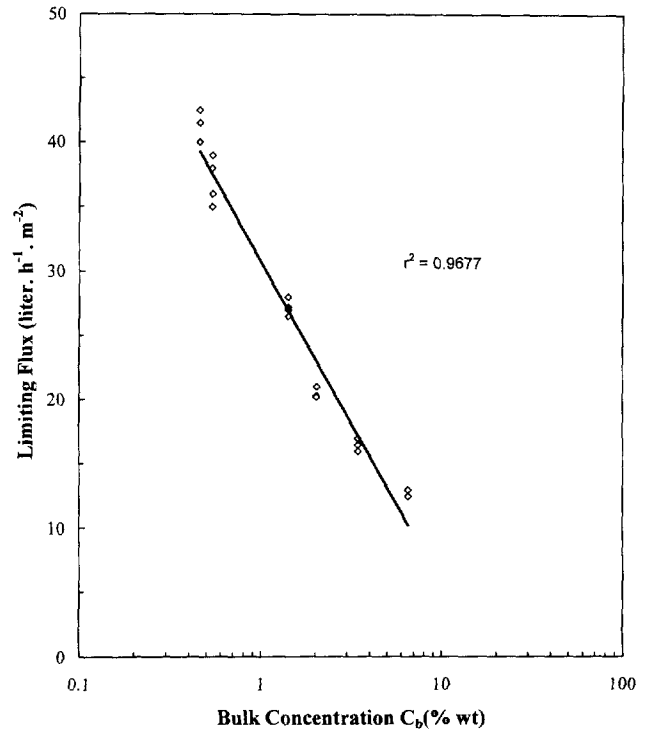


Fig. 3. Variation of limiting permeate flux with bulk concentration of solute.

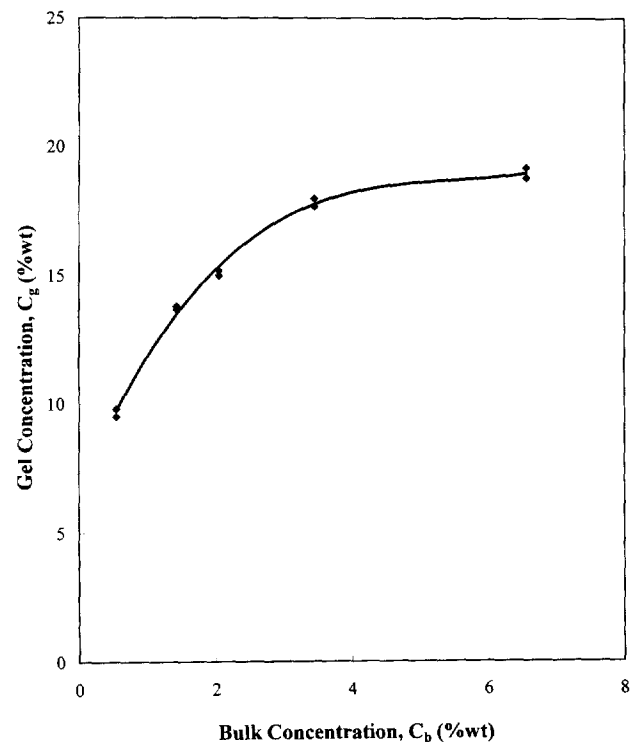


Fig. 4. Relationship between gel concentration C_g and bulk concentration C_b .

significant error, it can be assumed that the solute concentration in permeate is zero, thus justifying the validity of Eq. (1). As shown in Fig. 3, forcing a straight line through the data points is possible, but only with $r^2 = 0.968$. In this case, it is rather difficult to identify the actual values of the limiting flux as indicated by the scatter of the data points, thus supporting the earlier observation that there was no constant limiting flux value. For this reason, deviation from the linear model is to be expected. A possible reason for the observed linear relationship in Fig. 3 was due to the narrow range of C_b investigated. A non-linear relationship would have been shown if the range of concentration investigated was wider.

With the experimental set-up used in the investigation, it was possible to actually determine the steady state gel layer concentration, C_g , which is equal to the dry weight of the gel itself. As indicated by Eq. (1), the gel layer concentration should be independent of the bulk concentration. However, as shown in Fig. 4, it is obvious that C_g depends strongly on C_b and is not a constant value as predicted by the gel polarisation model. There were indications by others [9] that C_g is not a constant but depends on C_b and the crossflow velocity. In addition, different authors have reported different values of C_g for different solutes.

4. Conclusion

In comparing the observed results with those predicted from the gel model, it is evident that the model cannot be applied to the clarification of starfruit juice. The main reason for the deviation is believed to be due to the high content of pectin in the sample.

5. Nomenclature

C_g	Gel layer concentration of solute
C_b	Bulk concentration of solute (equal to feed concentration)
D	Diffusion coefficient
J	Flux
J_∞	Limiting flux
k	Mass transfer coefficient
R_m	Hydraulic resistance
δ	Boundary layer thickness

Acknowledgements

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References

- [1] D.A. Heatherbell, J.L. Short, P. Strübi, *Confructa* 22 (1977) 157.
- [2] D.E. Kirk, M.W. Montgomery, M.G. Kortekaas, *J. Food Sci.* 48 (1983) 1663.
- [3] E.L. Wilson, D.J.W. Burns, *J. Food Sci.* 48 (1983) 1101.
- [4] S.S. Koseoglu, J.T. Lawhon, E.W. Lusas, *Food Technol.* 45 (1991) 124.
- [5] M. Mulder, *Basic Principles of Membrane Technology*, Kluwer Academic Publishers, The Netherlands, 1991.
- [6] M. Cheryan, *J. Food Process Eng.* 1 (1977) 269.
- [7] O. Omosaiye, M. Cheryan, *J. Food Sci.* 44 (1979) 1027.
- [8] W. Pilnik, A.G.J. Voragen, Pectic substances and other uronides, in: A.C. Hume (Ed.), *The Biochemistry of Fruits and their Products*, Academic Press, London, 1970.
- [9] S.I. Nakao, T. Nomura, S. Kimura, *AIChE J.* 25 (1979) 615.