



## XVII<sup>th</sup> World Congress of the International Commission of Agricultural and Biosystems Engineering (CIGR)

Hosted by the Canadian Society for Bioengineering (CSBE/SCGAB)  
Québec City, Canada June 13-17, 2010



### SEPARATION OF BENZOIC ACID FROM CRANBERRY JUICE BY USE OF NANOFILTRATION MEMBRANES

DAT QUOC LAI<sup>1,2</sup>, NOBUHIRO TAGASHIRA<sup>2</sup>, SHOJI HAGIWARA<sup>1</sup>, MITSUTOSHI NAKAJIMA<sup>3</sup>, TOSHINORI KIMURA<sup>4</sup>, HIROSHI NABETANI<sup>1</sup>

1 D.Q. LAI, National Food Research Institute, NARO, 2-1-12 Kan-nodai, Tsukuba, Ibaraki 305-8642 Japan, nabetani/quocdatl/shoji/@affrc.go.jp

2 N. Tagashira, Graduate School of Agricultural Science, Hokkaido University, Kita 9, Nishi 9 Kita-ku, Sapporo 060-8589 Japan, Aohata Corporation, 1-1-25 Tadanouminaka-machi, Takehara City, Hiroshima 729-2392 Japan, ntagashi@orange.ocn.ne.jp

3 M. Nakajima, Graduate School of Life and Environmental Sciences, University of Tsukuba, 1-1-1 Ten-noudai, Tsukuba, Ibaraki 305-8577 Japan, nakajima.m.fu@u.tsukuba.ac.jp

4 T. Kimura, Graduate School of Agricultural Science, Hokkaido University, Kita 9, Nishi 9 Kita-ku, Sapporo 060-8589 Japan, toshibio@bpe.agr.hokudai.ac.jp

#### CSBE100661 – Presented at Symposium on Nanotechnologies Applied to Biosystems Engineering and the Environment Conference

**ABSTRACT** Benzoic acid has been used widely in food industry and cosmetic industry as a preservative because of its antibacterial activity. Cranberry is rich in benzoic acid, about 100 ppm in the cranberry juice (10 °Brix). The concentration implies that if the excess benzoic acid in cranberry juice can be recovered at low cost, it will be a promising natural preservative. In this study, efficient conditions of nanofiltration for benzoic acid recovery from cranberry juice were investigated. Fifteen kinds of commercial nanofiltration membranes were tested with membrane test cell (C60F, Nittodenco, Japan) for checking benzoic acid separation ability from cranberry juice. Seven kinds of membrane (HC50, NFT50, G5, Desal-DK, DRA4510, UTC60 and NTR7250) showed relatively low rejection (less than 50 %) against benzoic acid while rejection against other components such as sugars, organic acids (citric acid, quinic acid and malic acid) and anthocyanins was high (more than 70 %). The effect of pH on the nanofiltration efficiency was also investigated. When pH increased from 2.5 to 4.5, the benzoic acid rejection decreased and negative rejection was observed with some membrane, while the rejection against other components was still high and increased with pH. Therefore, high separation efficiency was obtained at pH 4.5. The nanofiltration conditions for benzoic acid recovery from cranberry juice were clarified in this study.

**Keywords:** Nanofiltration, benzoic acid, cranberry juice, pH effect, negative rejection

**INTRODUCTION** Benzoic acid ( $C_6H_5COOH$ , molecular weight = 122,  $pK_a = 4.19$ ) has been known as a preservative in food industry from 1875 because of its antimicrobial activity (Samson *et al.*, 2004). It is often used in combination with other preservatives and, on the basis of its higher activity at acidic pH in sour foods such as jellies, marmalades, fish preserves, margarine paste and so on. Benzoic acid naturally occurs in some fruits and spices, especially in cranberry (*Vaccinium macrocarpon*) (Belitz *et al.*,

2009). The content of benzoic acid in cranberry is 4.741 g/kg fresh weight, including about 10 % in free state and 90 % in bound state (Zou *et al.*, 2002). In 2008, the production of cranberry in USA was approximately 393,000 tons, which is approximately 90 % of the total production of the world (source: NASS/USDA). Therefore, it is promising to recover benzoic acid from cranberry profitably to use as a natural preservative.

Nanofiltration (NF) is a pressure driven membrane process with the molecular weight cut off (MWCO) situated between ultrafiltration and reverse osmosis. The main advantage of nanofiltration is that it can be operated at the low temperature. Therefore, the energy consumption is low and the thermally sensitive components can be retained. Besides, the nanofiltration system is also simple to install and to operate. Nowadays, there are many literatures which reports the research and the application of membrane technology in food industry such as: fractionating sugars (Goulas *et al.*, 2002), recovering and purifying amino acids (Kovacs and Samhaber, 2009), recovering and purifying the fermentation products (Li *et al.*, 2008), concentrating coffee extract (Vincze and Vatai, 2004), concentrating juices (Warczok *et al.*, 2004) and removing the salt in whey protein production (Van der Horst *et al.*, 1995).

In nanofiltration, separation is achieved by the charge exclusion and the size exclusion. The size exclusion (sieving effect) depends on the membrane structure: a denser structure leading to a lower permeation. The charge exclusion of ions depends on the charge of the membrane, the ionic strength and the valence of ion: the latter two influence the charge density and the iso-electric point of the membrane (Nilsson *et al.*, 2008). Therefore, it can be stated that NF can be useful to separate both charged solutes and neutral solutes (Hussain *et al.*, 2007).

The present study focuses on the feasibility of the application of nanofiltration to separate benzoic acid from the cranberry juice. Firstly, the study is investigated on the effect of types of commercial membranes, which is aimed to choose the suitable membranes for the separation. Secondly, the effect of pH of the cranberry juice on the separation is investigated to determine a suitable value for the benzoic acid recovery.

## MATERIALS AND METHODS

**Materials** The concentrated cranberry juice (50 °Brix) was supplied by Aohata Company. In our research, in order to recover benzoic acid, the concentrated juice was diluted by deionized water to reach 10 °Brix. All reagents were supplied by Wako Pure Chemical Industries, Ltd (Japan) with the analytical grade.

**Membrane apparatus** The flat-membrane test cell C60F supplied by Nitto Denko Corporation (Japan) was operated in the batch mode (Fig. 1). The experiments were conducted under nitrogen atmosphere and the operating pressure was adjusted by connecting to the nitrogen cylinder. The membrane cell was placed on the magnetic stirrer and the agitation was provided by the magnetic spin bar installed inside the cell. The membranes were cut into circular discs (7.5 cm diameter and 32 cm<sup>2</sup> affective area) and placed in the membrane cell (on porous support sheet). The initial volume of the feed in the cell was 200 mL. In order to eliminate the concentration polarization on the surface of the membrane, the velocity of the stirrer was set at 500 rpm.

**Table 1:** The characteristics of membranes

Manufacturer	Type	NaCl rejection (%)	pH rank	Temperature rank (°C)	Pure water permeability (L/m <sup>2</sup> .h.bar)	Material
DDS	HC50	40 ~ 60	2 – 10	<60	6.24	Composite
	NFT50	55	2 – 10	-	10.76	Composite
Desalination systems	G5	1000*	2 – 11	<50	1.33	Composite
	G-10	2500*	2 – 11	<50	4.75	Composite
	Desal – DK	50	2 – 11	<50	3.90	Polyamide
	Desal – DL	15	2 – 11	<50	6.90	Polyamide
KOCH	MPF 34	35	0 – 14	<70	1.39	Polysulfone
	MPF 44	25	2 – 10	<40	0.94	Polyacrylonitrile
	MPF –50	700*	4 – 10	<40	0.5	PAN
	MPF –36	10	0 – 13	<70	31.16	Polysulfone
Daisen	DRA 4510	45	2 – 11	-	4.96	Composite
Torai	UTC 60	55	-	-	5.70	Polyamide
Nitto Denko	NTR 7430	30	2 – 11	<90	22.63	Sulfonatepolyethersulfone
	NTR 7450	50	2 – 11	<90	11.78	Sulfonatepolyethersulfone
	NTR 7250	60	2 – 8	<60	-	Polyvinyl alcohol

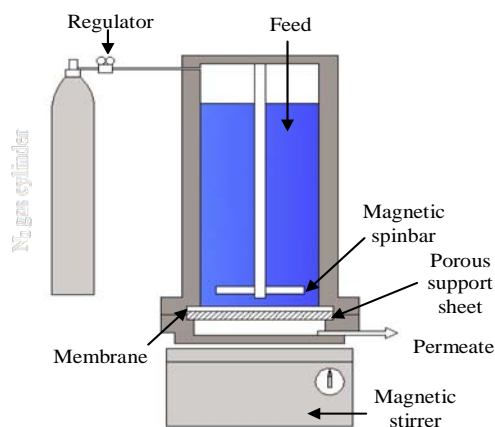
Source: from the manufactures

(\*): based on the molecular weight cut off

**Membranes** Fifteen different kinds of membranes were supplied from 6 manufacturers. The characteristics of the membranes were shown in Table 1. With each experiment, the fresh membranes were soaked in deionized water for at least 48 h before use.

**Experimental procedure** At first, to evaluate the feasibility of nanofiltration to recover benzoic acid, the cranberry juice (10 °Brix) was pretreated by ultrafiltration with GR61PP membrane (MWCO: 20000 Da) to remove macromolecules. The conditions of ultrafiltrations were as follows: operating pressure was 20 bar and the temperature was 27 °C. The pH value of the pretreated cranberry juice was 2.5. Fifteen kinds of commercial nanofiltration membranes which were supplied by six manufacturers were tested with the following parameters: operating pressure was 30 bar, and the temperature was 27 °C. Then, to evaluate the effect of pH on the separation of benzoic acid, the cranberry juice (10 °Brix) without pretreatment was filtrated by suitable nanofiltration membranes selected from the first run with following parameters: the range of pH was 2.5 – 4.5, operating pressure was 30 bar and the temperature was 27 °C. The pH of the cranberry juice was adjusted by NaOH.

**Analytical methods** The contents of glucose and fructose were analyzed by using YMC-Pack Polyamine II (250x4.6 mm ID) (supplied by YMC Co. Ltd, Japan) and Waters 717 plus Autosample (with Waters 515 HPLC pump) coupled to a refractive index detector. The column was maintained at 35°C and the mobile phase was acetonitrile/water (70/30) at the flow rate of 1 mL/min.



**Figure 1:** Scheme of the flat membrane test cell C60F

The contents of benzoic acid were analyzed by using YMC – Pack Pro C18 (150 x 4.6 mm ID) (supplied by YMC Co. Ltd, Japan) and the Waters Alliance 2695 HPLC system coupled to Water 2487 UV detector (UV 210 ~ 240). The column was maintained at 45 °C and the mobile phase was methanol/sodium phosphate buffer pH 4.5 (25/75) at the flow rate of 0.9 mL/min.

The contents of quinic acid, malic acid and citric acid were analyzed by the capillary electrophoresis system (Agilent G1600A) with a fused silica capillary (l=72 cm, L = 80.5, ID = 50 µm) coupled to the detector: Sig. = 350/20 nm, Ref. = 275/10 nm). The buffer was Agilent plating bath buffer for CE (part No. 5064 – 8236) and the injection was done at 6 s x 50 mbar. The capillary was maintained at 15 °C. The voltage was -25 kV.

The contents of anthocyanin were determined indirectly by measuring the absorbance of the cranberry juice with the spectrophotometer set at the wavelength of 530 nm.

**Performance parameters** The performance of the membrane process was expressed in terms of permeate flux (L/m<sup>2</sup>.h) and the rejection of benzoic acid, sugars (being defined as the sum of glucose and fructose), organic acids (being defined as the sum of quinic acid, malic acid and citric acid) and anthocyanin.

The rejection ( $R_o$ ) was calculated from the following formula:

$$R_o = 1 - C_p / C_f$$

The  $C_p$  and  $C_f$  were concentrations of the solutes in the permeate and the feed, respectively. The  $R_o$  and permeate flux were determined when the permeate volume reached at 10 – 20 mL.

## RESULTS AND DISCUSSION

**The capacity of nanofiltration to recover benzoic acid from the UF pretreated cranberry juice** The performance of separation by the nanofiltration membranes was shown in the Fig. 2. The observed rejection of benzoic acid was the lowest in all the membranes. The  $R_o$  of benzoic acid was approximately 30 % or lower (except NTR7250, MPF44 and MPF34). No one exceeded 55 % whereas the  $R_o$  of sugars, organic acids and anthocyanins were around 70 % or higher (except G10, MPF44, MPF50, MPF36, NTR7430 and NTR7250). The difference of the  $R_o$  between benzoic acid and others was not lower than 20 % (except the membranes made by KOCH). Some membranes such as HC50, NFT50 and DRA4510 made the difference exceeding 50 %. This phenomenon could be explained by the difference of the molecular weights. In the membrane with the given pore size, the smaller the molecular weight was, the higher the permeability was. The molecular weight of benzoic acid was the smallest when compared with sugars, other organic acids and anthocyanins. It meant that the molecular size of benzoic acid was smaller than any of the other compounds. Consequently, the permeability of benzoic acid through the membranes was the highest. Except MPF36, the  $R_o$  of anthocyanins was also the highest because of the high molecular weight.

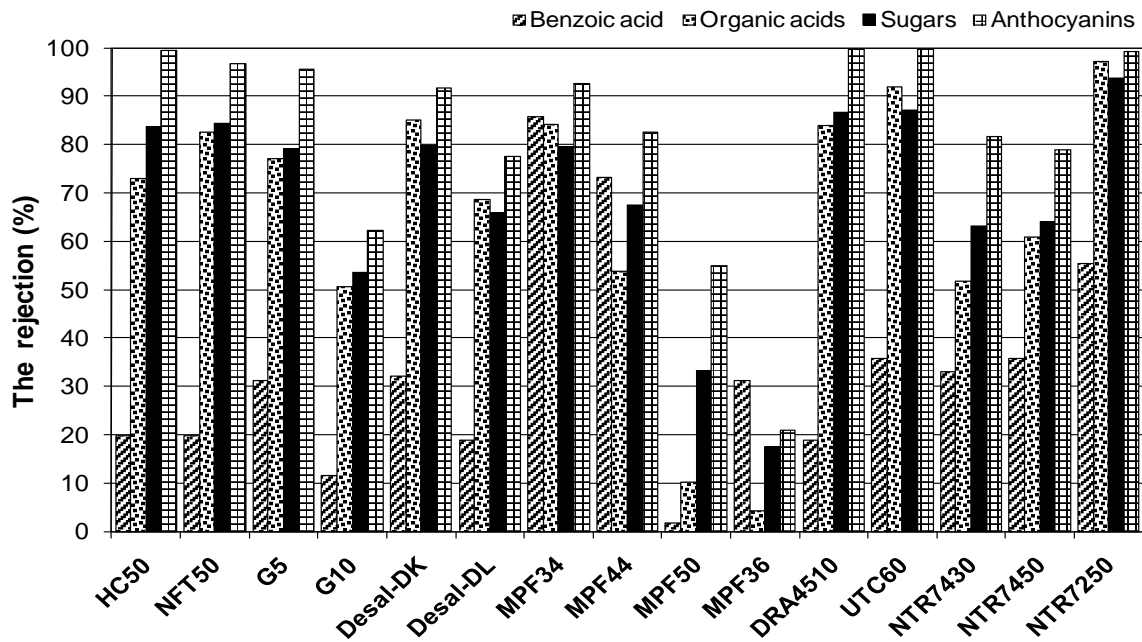


Fig. 2: Rejection against components in the cranberry juice

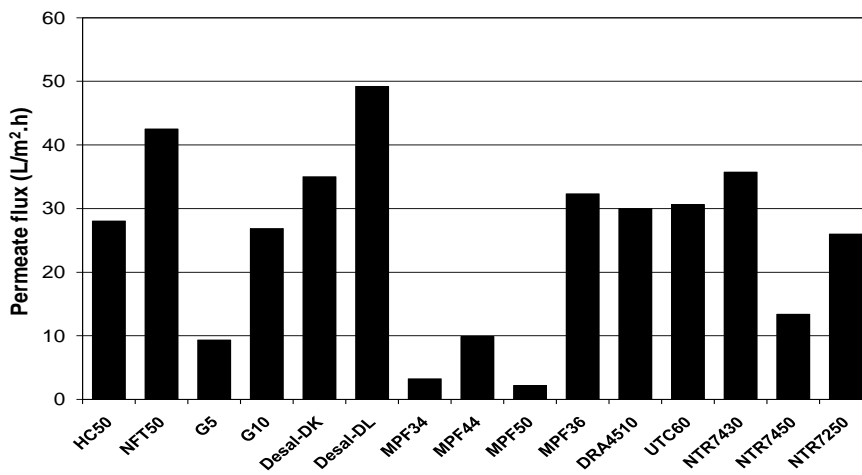


Fig. 3: The permeate flux of nanofiltration for the cranberry juice.

The Fig. 3 shows that the permeate flux depended on the structure of the membrane. The permeate flux of the membranes was significantly different, even in the case the pore sizes of the membranes were the same (for example, NFT50 and UTC60). Comparing the pure water permeabilities in Table 1, it was clear that the denser the membrane was, the lower the permeate flux became because the denser membrane resisted permeation more significantly.

Based on the differences between benzoic acid and other components in the cranberry juice, the suitable membranes to separate benzoic acid from other components were chosen and the effect of pH on the separation was evaluated. The criterion of the

selection of the membrane was that the difference in rejection should be approximately or higher than 40 %. There were 7 kinds of membranes, such as HC50, NFT50, G5, Desal-DK, DRA4510, UTC60 and NTR7250, which were promising to separate benzoic acid.

### The nanofiltration to recover benzoic acid from the untreated cranberry juice

The effect of pH on the sugar rejection in the untreated cranberry juice The Fig. 4 shows that the effect of pH on the sugar separation was not significant. The change in the sugar rejection obtained by NFT50, Desal DK, DRA4510 and UTC60 were insignificant (below 5%) when pH was adjusted to 2.5 – 4.5. However, a significant effect of pH on the glucose rejection was observed in the membrane Desal G5 and NTR7250.

To explain this phenomenon, the mechanism of the solute permeation through the nanofiltration membrane should be investigated. The permeation of the solutes through the membrane was affected by the convection, the diffusion and the migration (Szymczyk *et al.*, 2003). As the sugars are neutral, the main permeation through the membrane should have been caused by the convection, therefore, more significantly, affected by the structural change. The pH can influence the charge on the surface and the pore wall of the membrane which lead to the swelling because of the electrically repulsive force. The more the intensity of the repulsive interaction was, the more the swelling was (Mänttari *et al.*, 2006; Izák *et al.*, 2007). As the swelling of the membrane could cause the increase in a pore size, it could also affect the retention of solutes on the retentate side. The isoelectric point (pI) of most membranes made from polyamide ranked from 4 – 6 and depends on the ratio of the number of amine groups to that of carboxyl groups (Mänttari *et al.*, 2006). However, in the cranberry juice, the presence of calcium (approximately 75 ppm) could make pI change to the neutral value. For instance, in the case of NFT50, the isoelectrical point was 4.2- 4.3, and exceeded 5.5 with the presence of CaCl<sub>2</sub> (1 mM) (Teixeira *et al.*, 2005). The reason for this was because the negatively charged group in the membrane adsorped Ca<sup>2+</sup>. As the adsorption made the negatively charged group reduce, it made pI change to the neutral value.

Consequently, in our study, even at pH 4.5, most investigated membranes could be positively charged or very slightly negatively charged. As the increase in pH from 2.5 to

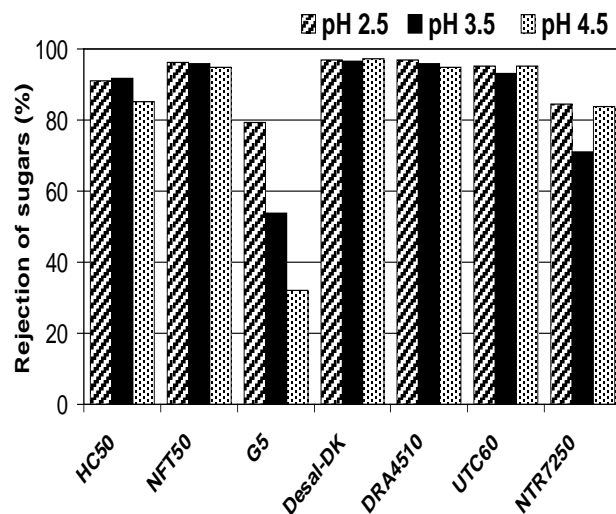


Fig. 4: The effect of pH on the sugar rejection during nanofiltration of the cranberry juice

Table 2: The sodium content in the cranberry juice and the permeate (ppm)

	pH 2.5	pH 3.5	pH 4.5
Cranberry juice	75	2900	5900
HC50	14	1740	2350
NFT50	15	1640	1410
G5	42	3080	5110
Desal-DK	8	1340	620
DRA4510	30	1130	1440
UTC60	7	1210	980
NTR7250	24	2570	2370

4.5 decreased the intensity of the charge in the membranes, the swelling due to the electrical repulsion was reduced and the pore size became smaller.

Besides that, the presence of an inorganic ion could also affect the swelling of the membrane which could influence the rejection (Nilsson *et al.*, 2008; Mandale and Jones, 2008). In our study, when pH of the cranberry juice increased, the sodium content also increased because pH was adjusted by NaOH. The sodium content in the cranberry juice at pH 2.5, 3.5 and 4.5 was 75 ppm, 2900 ppm and 5900 ppm, respectively. Consequently, the increase in pH could make the pore size larger when the sodium content increased because of the swelling as stated above.

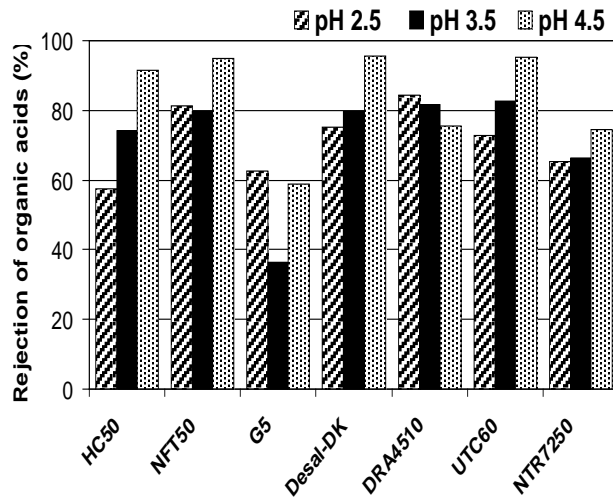
The change in the sugar rejection could be explained by the decrease in the density of charge and the increase of the sodium content when pH in the cranberry juice increased. The former made the active layer of the membrane denser and the latter made it more expanded. In addition, the change in the charge of the membranes could affect the permeation of sugar through the membranes because sugars were permanently polar. The contribution of these factors to the effect of the rejection depended on the material of the membrane and on the ratio of the molecular size to the membrane pore size. In the case of G5 and NTR7250, as the material could be more susceptible to the change in pH and to the sodium content in the cranberry juice than the materials of other membranes or as the size ratio was low, the change of the rejection of sugar was clearer. In the other membranes, since the size ratio could be high, the sugar rejection was not clearly affected by the pH value and still stable at such a high value (over 90 %). This hypothesis was reinforced when the rejection of anthocyanins was observed. The molecular size of anthocyanins was larger than glucose and fructose. Therefore, the molecular size of anthocyanins should have been higher than the pore size of the membrane. Consequently, the rejection of anthocyanins was higher (approximately 100 %) and not affected by the change in pH of the cranberry juice.

The effect of pH on the organic acid rejection in the unpretreated cranberry juice  
From the data in Fig. 5, it was clear that the organic acids rejection trended to become higher with the increase in pH (except G5 and DRA4510). There were some effects influencing the rejection of organic acids. Not only could the pore size be changed by the change in pH and the sodium content as stated above, but also the dissociation and the change in the molecular size also affected the change of the rejection. Because the charge of the membrane could still be positive even at pH 4.5, the dissociated organic acids could move through the membranes more easily than the undissociated ones. However, the molecular size of the dissociated acids became larger because the hydration made the Stokes radius larger. It made the retention of organic acids on the retentate side higher. Therefore, those were some reasons why the rejection of organic acid changed when pH of the cranberry juice changed. In the case of HC50, NFT50, Desal-DK, UTC60 and NTR7250 membranes, when pH value became higher, the causes to have increased the rejection became more predominant. Consequently, the rejection of organic acid increased.

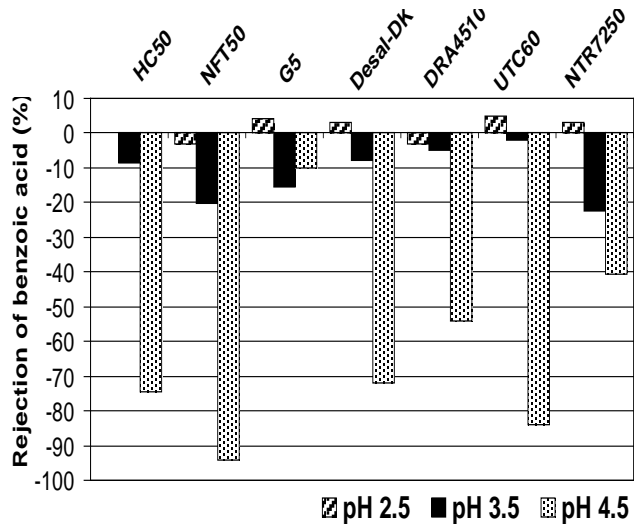
The effect of pH on the benzoic acid rejection in the untreated cranberry juice The rejection of benzoic acid was strongly affected by pH (Fig. 6). The rejection of benzoic acid decreased according to the increase in pH with all the investigated membranes. For examples, the rejection of benzoic acid by NFT50 at pH 2.5, 3.5, 4.5 were -3 %, -20.4 % and - 94.1 %, respectively. With the Desal-DK5 membrane, the rejections were 3 %, - 7.8 %, -72 %, respectively. The negative rejection proved that the diffusion and the migration of electrolyte considerably contributed to the passing through the membrane. Besides the effects as stated above, there were some more attributes which should have been investigated. Compared with other organic acids in the cranberry juice, the molecular size of benzoic acid was the smallest. Therefore, it could pass the pores and move through the membranes more easily. The passing was reinforced when benzoic acid was dissociated because of the electrical interaction between the positively charged membrane (in the case  $\text{pH} < \text{pI}$ ) and the negatively charged dissociated benzoic acid.

Simultaneously, when the sodium concentrations in the cranberry juice increased with pH, its concentration in the permeate size also increased (Table 2). Therefore, the passing of the dissociated benzoic acid through the membrane to neutralize the positive charge in the permeate size increased (Donnan, 1995). Consequently, the rejection of benzoic acid reduced and became negative when pH in the cranberry juice increased. The negative rejection of benzoic acid indicated that benzoic acid can be recovered from cranberry juice by nanofiltration.

The effect of pH on the permeate flux The effect of pH on the permeate flux is shown in the Fig.7. Except Desal-DK membrane, the increase in pH value of the cranberry juice made the permeate flux decrease. The effect of pH on the permeate flux was explained by the change in the hydrostatic resistant due to the swelling, the electroviscous effect and the osmotic pressure. Firstly, as the swelling made the membrane become looser, the resistant became lower and the permeate flux became higher. Secondly, the effect of pH on the permeate flux was related to the electroviscous effect, which was a physical phenomenon that occurred when an electrolyte solution was



**Fig. 5:** The effect of pH on the organic acids rejection during nanofiltration of the cranberry juice



**Fig. 6:** The effect of pH on the benzoic acid rejection during nanofiltration of the cranberry juice



pressed through a narrow capillary or a pore with a charged surface (Childress and Elimelech, 2000). The increase in ion strength increased the augmentation of the charge on the pore surface and decreased the passing of water through the membranes because of this effect. Finally, the increase in pH caused the increase in sodium (Table 2). As the augmentation in the sodium content increased the osmotic pressure, the transmembrane pressure reduced. Consequently, the permeate flux decreased because of the increase of the osmotic pressure. In our study, except Desal-DK, the factors, especially the increase of the osmotic pressure, which made the permeate flux decrease were predominant, it could be stated that the permeate flux decreased with the increase in pH in the cranberry juice. In the case of the Desal-DK membrane, the effect of pH on the pure water permeability was observed and it was not significant. Maybe, the effect of the sodium content on the swelling was predominant and consequently, the permeate flux was the highest at pH 4.5.

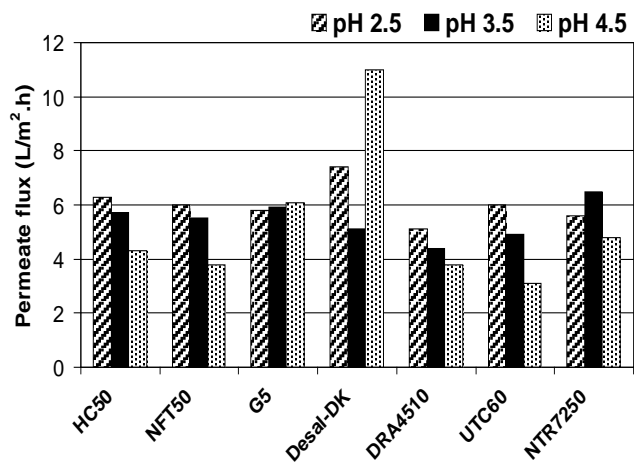


Fig. 7: The effect of pH on the permeate flux during nanofiltration of the cranberry juice

**CONCLUSION** This paper has investigated the feasibility of the application of nanofiltration to recover benzoic acid from the cranberry juice. The performance of the nanofiltration membranes and the effect of pH on the nanofiltration were observed. The rejection of benzoic acid by nanofiltration was significantly different from other components in the cranberry juice such as anthocyanins, sugars, other organic acids and minerals. The performance of the nanofiltration to recover benzoic acid from the cranberry juice depended on the material and the structure of the membranes which were provided from different manufactures. It was also influenced by the pH of the cranberry juice. The rejection of benzoic acid could become lower than -40 % at pH 4.5. With the NFT50 membrane, it could become -94 %. The results of this study indicated that the nanofiltration was a promising method to recover benzoic acid from the cranberry juice. More work would be required to evaluate the feasibility of a pilot scale and to determine the optimal parameters for nanofiltration more precisely and accurately.

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