# Optimal clarification of apple juice using crossflow microfiltration without enzymatic pre-treatment under different operation modes

**Fawad Khan[1], Waheed Ur Rehman[1], Mohammad Younas[1], Amir Muhammad[1], Wajid Ali[1], Imran Khan Swati[1]**

[1]Department of Chemical Engineering, University of Engineering and Technology, Peshawar, P.O Box 814, Pakistan

*e-mail: m.younas@uetpeshawar.edu.pk

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## Abstract

In this study, appropriate operating values for the clarification process were identified by investigating the influence of temperature, transmembrane pressure (TMP) and feed flow rate on the crossflow microfiltration (CMF) process without enzymatic pre-treatment. Multichannel ceramic membrane with active filtration area 0.085 $m^2$ and nominal pore diameter of 0.2μm was used in lab-scale microfiltration unit to attain clarified apple juice. The experiments were performed using two modes, total recirculation mode and concentration mode. In total recirculation, the performance of flux with time was observed at feed flowrate 11 $L.min^{-1}$ and at TMP of 3 and 4 bar respectively. A reduction in permeate flux was observed with time. The results showed in contrast to 3 bar run, the flux values at 4 bar was higher but the decay in flux of permeate with time at 4 bar was more rapid. In the concentration mode, albeit flux behavior was nearly the same as in total recirculation mode; the values of flux were much lower in concentration mode. The CMF had a noticeable impact on the physiochemical properties of apple juice, such as reduction in viscosity, turbidity and soluble solids. However, no change was noticed in the pH of apple juice. The study showed that CMF process is suitable for clarification of apple juice. The product obtained can be applied further to juice processing.

**Keywords:** clarification, crossflow microfiltration, multichannel ceramic membrane, apple juice, permeate flux

## Abbreviations

| | |
|---|---|
| CMF | Crossflow n |
| ºBrix | gram of sucrose per 100gram of solution |
| TMP | Transmembrane Pressure |
| TSS | Total soluble solids |
| NTU | Nephelometric Turbidity Unit |
| cP | centipoise |
| UPVC | Unplasticized polyvinyl chloride |

## Introduction

Currently, a considerable variety of new products made from clarified juice, have emerged in the market. The two principle features of these products are transparency and homogeneity, which can be attained by the entire removal of all the suspended particles. A few of these products are sparkling clear beverages, pastries, uniform pulpy fruit blend, and cosmetic products. The broad list of products indicates the existence of numerous opportunities in the market for the clarified juices made from fruits or vegetables. In addition to these markets, a demand for high quality juice also exist [1-3]. Unfortunately, the classical clarification and stabilization processes use chemical and thermal treatments that significantly deteriorate the quality of juice. Furthermore, the traditional clarification process involves several steps (i.e depectinization, centrifugation, addition of finning and filtration agents) which are tedious and time consuming [4]. Membrane technology provides a technological solution to the problems encountered in classical clarification and stabilization processes. Particularly cross microfiltration (CMF) is valid alternative for these classical processes as it provides a non-thermal approach and avoids the use of chemicals [5]. Other advantages of CMF include small processing time, low energy consumption and it is a single step continuous clarification process. MF is able to retain suspensions, macromolecules and bacteria due to its small pore size ($\Theta \leq 0.2\mu m$) thus ensures clear (transparent and homogenous) [6] and microbiologically stable [7] juice without deteriorating its natural aroma and colour. The performance of CMF is notably affected by the pulp content present in fruit juices. The pulp content forms a fouling layer on membrane surface [8] which results in permeate flux reduction. Fruit juices are treated with enzymes before subjecting it to CMF in order to reduce viscosity of the pulp content and fouling of membrane. However, rapid reduction in permeate flux still occurs [9], resulting in an ineffective apple juice clarification which limits the commercial use of CMF in juice processing. Moreover, the cost of membrane replacement and enzymatic pre-treatment makes the clarification process more expensive. The manufacturers need to keep lower prices of apple juice in order to compete in the market. The expensive clarification process adversely affects their profit pushing them to reduce the cost of production line and enhance efficiency [10]. The enzymatic hydrolysis pre-treatment process is one the reasons for high running cost of production line. Previously, studies on membrane based clarification of apple juice have utilized enzymes to aid filtration [2, 11-13, 14]. However, unsatisfactory permeate flux was reported by these studies. The aim of this study was to evaluate the CMF behaviour of apple juice without pre-treatment enzymatic hydrolysis.

## Material and Methods

### Apple Fruit Juice

Fresh apples were bought from the local market in Peshawar and a feed juice was obtained as under:
The apples were thoroughly washed with tap water, peeled, cut and deseeded with a stainless steel knife and were then grinded into a single strength juice by centrifugal shredder (MJ-0176P Panasonic Multifunctional juicer/blender). The coarse particles were removed from the juice by sieving it through a slit screen. 0.6 g/L potassium metabisulphite was added to the juice as a

preservative. The feed juice was finally subjected to CMF to acquire a clarified juice.

## Membrane Setup and Procedures

A multichannel tubular ceramic membrane module (for specifications see Table 1), a pump (0.75 Hp, centrifugal) and control valves were incorporated in the CMF unit as shown in figure 1. The pressure measurements at the inlet and outlets of the membrane module were made by three pressure gauges. For the flowrate measurements, variable area flowmeters were installed at the feed and retantate line. The pressure and flow rate were regulated by three control valves installed at inlet and outlets of the membrane module.The temperature in the feed tank (capacity 10 L) was measured through a mercury filled thermometer.

The membrane module was washed with distilled water for trial runs. The feed apple juice was pumped into the CMF unit for clarification. To avoid any damage to equipment and juice quality, variations in TMP, feed temperature and flowrate were executed in suitable amplitude of operating conditions. Experimental trials were conducted in total recirculation and concentration mode. In total recycle mode, both permeate and retantate were supplied back to the feed tank, as a result the total volume remained constant. While in concentration mode, permeate was withdrawn and retantate was supplied back to the feed tank.

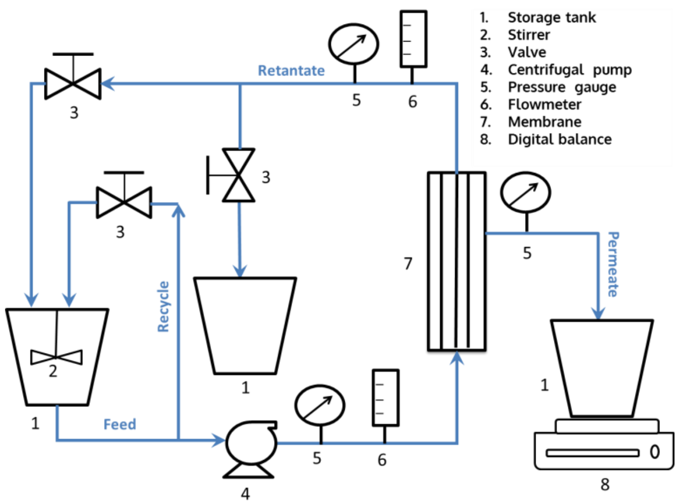


**Fig. 1: Laboratory scale Crossflow microfiltration unit.**

Pressure, flowrate and temperature were noted both at the inlet and outlet of the membrane module. Volumetric gain (V) in permeate with time (t) was also measured. The permeate flux (J) was calculated according to equation 1[15].

$$J = \frac{V}{A\,t} \tag{1}$$

Where

- $J$ is the permeate Flux ($Lm^{-2}hr^{-1}$)
- $V$ is the permeate volumetric gain (L)
- $A$ is the contact area of membrane( $m^2$)
- $t$ is the time flow of fruit juice (hour)

Equation 2 was used calculate TMP [16].

$$TMP = \left(\frac{P1 + P2}{2}\right) - P3 \tag{2}$$

Where

- $P1$ is the feed pressure (bar)
- $P2$ is the retantate pressure (bar)
- $P3$ is the Permeate pressure (bar)

**Table 1: Membrane and membrane module specifications**

| | |
|---|---|
| Manufacturer | Nanjing H&C Water Treatment Equipment Co., Ltd. China |
| Model | Model HCCM-200-40-19-06 |
| Dimensions (cm x cm) | 200 x 40 ø |
| Pore size (nm) | 200 |
| No. of channels (tubes) | 19 |
| Active surface area ($m^2$) | 0.085 |
| Membrane material | Aluminia, Zirconia oxide |
| Membrane housing | Unplasticized polyvinyl chloride (UPVC) |

The process of membrane cleaning was performed to increase the life span of membrane and to regain the permeate flux after each trial. Membrane cleaning procedure consisted of the following steps [17]: (1) rinsing the membrane with deionized water;(2) recycling 2% NaOH at 50°C for 15 minutes without filtration and 15 minutes with filtration;(3) rinsing with deionized water at 50°C until the pH is close to neutral (pH 7-8); and (4) sporadically adding 1% $HNO_3$ at 50°C.

## Analysis

Samples of Apple juice collected from permeate line of CMF experiments was used for quality analysis. The total soluble solids (TSS) content was directly determined in ºBrix (ºBx) with hand held portable refractometer (ºBrix: 0-80%, Min Div: ºBrix: 0.5%, accuracy: ºBrix: ± 0.5 %). The juice turbidity was assessed with Portable Hach 16800 turbiditimeter. The juice viscosity and pH values were determined with Oswald's capillary glass viscometer and digital pH meter.

## Results and Discussions

Effect of operating conditions in total recycle mode

### Effect of TMP

Permeate flux increased linearly with the increase in TMP up to 3 bar, while it remained at a constant plateau between 3 to 4 bar. It is evident from figure 2 that the highest permeate flux was achieved around 3 bar. The linear increase in permeate flux with TMP was due to the increase in driving force across the membrane [12]. Concentration polarization and cake layer formation [11] were responsible for forcing the permeate flux to remain at a constant plateau at high operating TMP. High operating pressure accelerated particle deposition on membrane surface and also compressed the deposited particles into thicker and denser fouling layer, thus high fouling resistance was offered [13]. The quality analysis of permeate showed that TSS increased from 12 to 14.5 ºBx. The removal of suspended particles dropped the turbidity to 1.7 NTU. The viscosity was reduced from 1.13 to 1.06 cP due to exclusion of pectin materials [14]. The pH value remained unchanged.

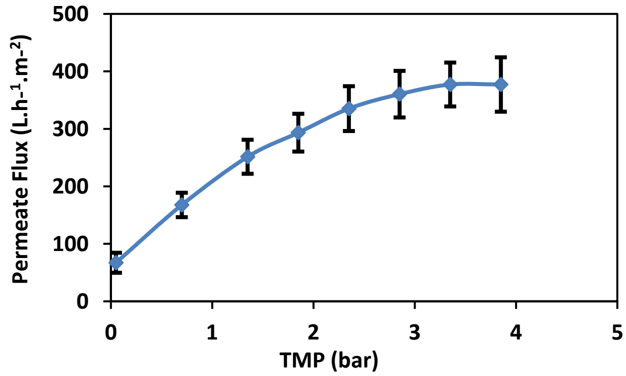


**Fig. 2: Influence of TMP on permeate flux (mean ± SD). Feed flow rate and temperature in Total recycle mode were kept constant at 20$^0$C and 11 L.min$^{-1}$ respectively (n=3 for each TMP).**

## Effect of Temperature

To illustrate the influence of temperature, the feed temperature was raised from 20°C to 40°C. Permeate flux increased with an increase in the feed temperature. The reduction in the viscosity and the increase in diffusion coefficient of macromolecules with rising temperature are two main factors responsible for higher permeation rate [18]. To minimize any thermal damage, the temperature was not raised any further [19]. Figure 3 represents the performance of permeate flux with feed temperature.

Feed temperature had a profound effect on the permeate quality parameters. With the rise in feed temperature, TSS increased to 14 ºBx while viscosity and turbidity dropped to 0.92 cP and 2 NTU respectively. However, the pH remained unchanged for feed temperature variations.

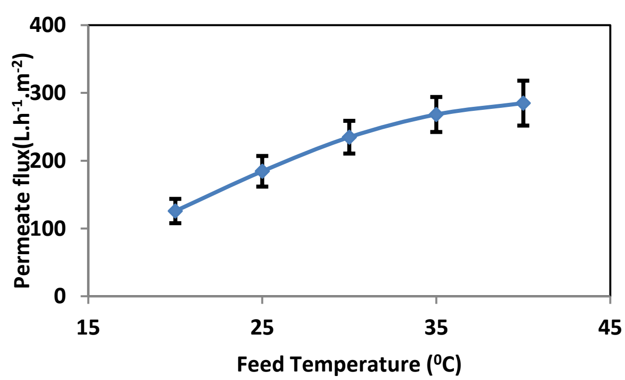


**Fig. 3: Effect of feed temperature on permeate flux (mean ± SD). Feed flow rate and TMP 3 Bar in Total recycle mode were kept constant at 11 L.min$^{-1}$ and 3 bar respectively (n=3 for each feed temperature).**

## Effect of Feed Flowrate

The TMP and feed temperature were fixed at 3 Bar and 20°C respectively. The permeate flux was enhanced from 125 to 440 L.h$^{-1}$.m$^{-2}$, when the feed flowrate was increased to 630 L.h$^{-1}$ as shown in figure 4. The crossflow velocity influenced the shear stresses at the membrane surface resulting in faster removal of the deposited particles over it [19, 20]. Therefore, the permeate flux enhanced with the increase in the feed flowrate.

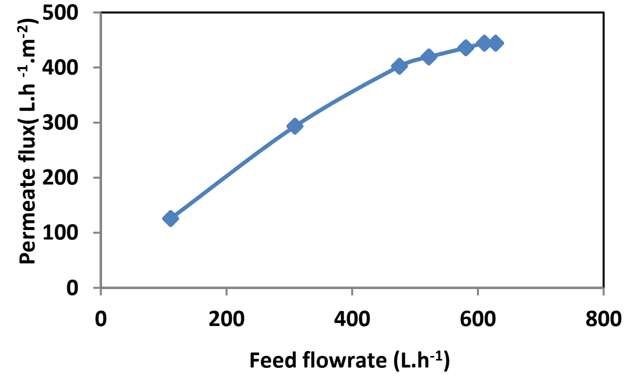


**Fig. 4: Effect of Feed flowrate on Permeate flux (Temperature: 20 ºC, TMP: 3 Bar) Total recycle mode**.

Permeate quality analysis was made for each incremental change in the feed flowrate. The TSS rose to 13.5 ºBx upon increasing the feed flow rate to 450 L.h$^{-1}$. Increasing the feed flow rate beyond this point did not affect the TSS anymore. Viscosity remained between 1 to 1.2cP for the feed flowrate variations. Turbidity was reduced to 1.7 NTU. No change was noticed in permeate pH.

## Effect of Operating Time

Two experimental trials were conducted at 3 and 4 bar as shown in figure 5. The feed flowrate was kept constant at 11 L.h$^{-1}$ for each trial. A gradual rise in permeate temperature was noticed from 20°C to 32°C with time course in the 3 bar. A sharp decrease in the permeate flux was observed in the initial 30 minutes of 3 bar run due to adsorption of colloidal material and growth of concentration polarization layer [21].

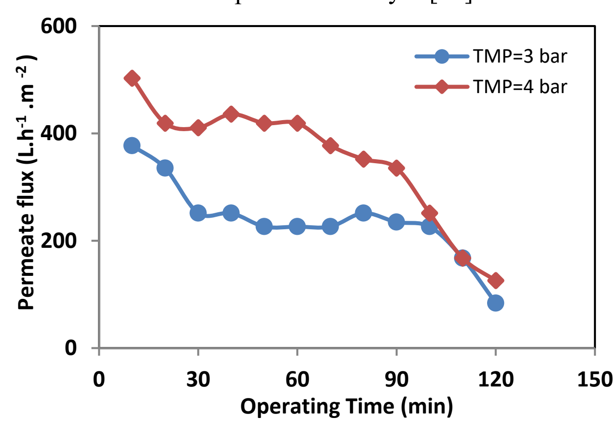


**Fig. 5: Effect of operating time on Permeate flux at different TMP (Feed flow rate: 11 L.h$^{-1}$.m$^{-2}$), Total recycle mode.**

Blockage of the membrane pores and cake layer formation over it [21] led to a steady decline in the permeate flux between 30 to 100 minutes. Afterwards, the permeate flux abruptly declined due to compaction of the fouling layer [22]. Flux decline pattern was similar at 4 bar. However, the flux values were higher than at 3 bar because of a high applied TMP. The temperature rise was also slightly higher at 4 bar because a high fouling resistance was offered for high operating TMP. Some problems are linked to a high operating pressure like accelerated flux decline and reduced duration of operating time [21]. It also compresses the deposited particles into a dense fouling layer which makes the membrane cleaning very difficult [22]. Initially at 3 bar, the TSS remained unaffected at 12 ºBx. With the formation of fouling layer and membrane pore blockage over time, the TSS was reduced to 11.5 ºBx. A further reduction in the TSS was not observed because of the diffusion of sugar molecules across the fouling layer. A similar pattern was observed at 4 bar. However, the values of TTS were much lower than at 3 bar. This is due to generation of a heavier and denser fouling layer at high TMP [21]. The permeate viscosity decreased initially in both runs and was then increased to 1 cP later on. A reduction in permeate turbidity was noticed in both trails. No change in pH was observed.

### Effect of operating time (concentration mode)

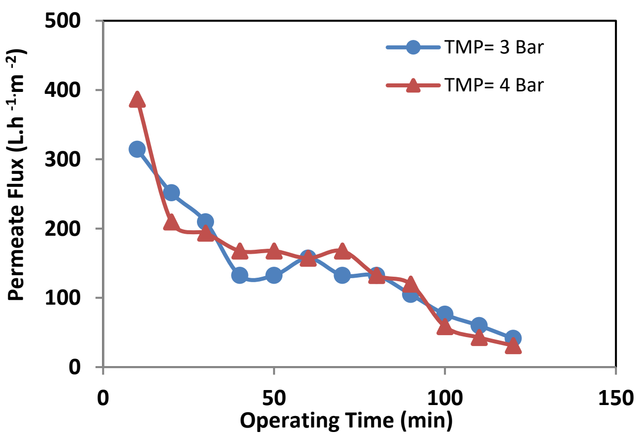


**Fig. 6: Effect of operating time on permeate flux of apple juice (Feed flow rate: 11 $L.h^{-1}.m^{-2}$), Concentration mode**.

Figure 6 shows flux-time curves carried out in concentration mode at 3 and 4 bar. The feed flow rate was held constant at 11 $L.min^{-1}$ for each run. Lower permeate flux values were achieved in concentration mode as compared to total recirculation mode. It was due to generation of extra resistance at membrane surface caused by accumulation of rejected particles [23]. The usual flux decline pattern was observed over time in each trail. Figure 6 depicts that the permeate flux rapidly declined in the beginning. The initial flux decline was caused by colloidal material adsorption and concentration polarization. However, the flux decline was sharper at 4 bar. Afterwards, the flux became stable and declined gradually due to pore blockage and formation of cake layer. At the end, cake layer compaction speeded up the permeate flux decline [22].

## Conclusions

The research analysis concluded that the CMF process is a reliable and advanced approach for apple juice clarification as it permits a simplified process while working at room temperature and boosts the juice quality by conserving its freshness and aroma. The permeation rate enhanced with the increase in TMP, feed temperature and flowrate. The permeate flux values obtained in total recirculation mode and concentration mode for 3 bar, 11 L/min and 30°C represents the optimum operating conditions. Therefore, under optimum conditions CMF in total recirculation mode assures a steady and elevated permeate flux.

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