Diafiltration based cow's milk partial demineralization - laboratory membrane filtration experiments

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## Abstract

Milk is the primary source of the nutrition mammals as it contains number of biological materials for the human body. These essential components are the proteins, lipids, vitamins and minerals. The large amounts of calcium and magnesium which can be find in the milk and have positive effect on the human health, can be extracted and used to enrich other food products with these components. Among the most common procedures for demineralization we can use ion exchange, nanofiltration and electrodialysis-based technologies. In this study the application of membrane filtration based milk partial demineralization and the statistical evaluations are detailed. The experiments were carried out using laboratory ultra- and nanofiltration units. The $2^P$ factorial experimental design was used for experimental planning. The statistical analysis of the results was carried out with the Statsoft Statistic program. Analyzing the influence of the operation parameters, the feasibility of the proposed technology was investigated. From the results of the membrane filtration experiments this technology is gentle and don't need to use extreme parameters and chemicals.

**Keywords:** Partial demineralization of milk, Statistical evaluation, Experimental Design

## *1. Introduction*

The milk is a complex lipid, carbohydrate and protein matrix which contains number of biological active components (Csapo, J., Csapone, K. Zs., 2002). Developments in the separation processes have created the opportunity for an entirely new approach to partial milk demineralization. Among the most common used processes for demineralization are based on ion exchange, nanofiltration and electrodialysis-based technologies. In this study membrane filtration (UF,NF) have been used for milk partial demineralization. The membrane is a selective barrier which can allows some components to pass through but other components cannot go. The membrane separation processes are selective and during the separation chemical changes not happen. In both processes was used cross-flow filtration where the solution flows under pressure along the membrane.

Depending on the pore size we can distinguished four type of membranes which are the microfiltration, ultrafiltration, nanofiltration and reverse osmosis (Mulder, Marcel 1996, Strathmann H., Giorno L et al., 2006, Fabry, Gy., 1995).

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The UF membranes are suitable to filter out macromolecules like proteins and polyphenol molecules. The pore size of an ultrafiltration membrane disposed between 10-100 nm, these pores are only permeable for the water, salts and the lower molecular weight components. The nanofiltration membranes pore size is between 1-10 nm, smaller than the ultrafiltration and microfiltration membranes. Nanofiltration membranes are widely used in the dairy industry for the partial demineralization of milk and whey (Fabry, Gy., 1995, Baker, L.A.; Martin 2007). The major purpose of this study is the partial demineralization of milk to decrease the monovalent ions from the matrix. To remove the monovalent ions complex membrane separation processes are used. In the first step (ultrafiltration) was the pre-concentration of milk where the bigger molecular weight components are retained. The next step was the diafiltration, the solution milk UF permeate was concentrated at a specified rate then the concentrate was diluted with a solvent and recycled to the system. To create an economical and operable technology it is important to find the optimal operation parameters. The $2^p$ factorial experimental design was used, the interactions between the measurements were disregard. The used $2^P$ experimental design contains a p factor which was measured in two level (minimum, maximum (Kemény, S., Deák, A., 1990).

## *2. Materials and methods*

### *2.1 Materials*

In this study one ultrafiltration and two nanofiltration membrane were used. The ultrafiltration membrane (Schumasiv 100nm) was used for milk pre-concentration. The next step was the permeate and the retentate diafiltration. Schumasiv type 5 nm pore size membrane for nanofiltration of the ultrafiltration retentate and the Membralox type 5 kDa membrane for diafiltration of the ultrafiltration permeate. The measured characteristic was the permeate flux, defined as follow.

- Flux

$$J = \frac{V}{A*t}$$

where:

J – permeate flux [L/(m2*h)],

V – permeate volume (L),

A - membrane surface ($m^2$)

t – filtration time (h)

*2.2 Methods*

*2.2.1 Experimental Design ($2^P$)*

In order to determine the optimal parameters for the membrane separation processes (pre-concentration, diafiltration) it is important to prepare the factorial experimental design. During the experimental design it is important to choose one maximum (+1) and one minimum (-1) value. Furthermore these combinations need to be the same quantity.

*2.2.2 Statistical method*

Statistical evaluation was performed using Statsoft Statistica program. During the experimental plans, the optimal transmembrane pressure and recirculation flow rate values was examined. In the next step the effect of the operational parameters were estimated, where the p and the $R^2$ value were the most important, because these numbers can show the effect of the two measured parameters (transmembrane pressure, recirculation flow rate) on permeate flux. The results are shown in the Pareto chart.

*3. Results and discussion*

*3.1 Experimental design ($2^P$)*

From the settings of the operation parameters and the results of experiments in case of milk ultrafiltration, presented in Table 1 is obvious that the optimal operation parameters, which shows the greatest flux value, are the fourth settings (32.17 l/$m^2$*h). The highest flux value was reached at 1.5 bar transmembrane pressure and 150 l/h recirculation flowrate.

From the results of the UF experiments shown in Table 1, it is clear that the transmembrane pressure and the recirculation flow rate significantly influenced the permeate flux.

Table 1. Experimental Design of Ultrafiltration (100nm UF)

| Sample | Transmembrane pressure | Recirculation flow-rate (l/h) | Flux (l/m2*h) |
|---|---|---|---|
| 1 | 1 | 100 | 23.73 |
| 2 | 1 | 150 | 29.02 |
| 3 | 1.5 | 100 | 25.6 |
| 4 | 1.5 | 150 | 32.17 |
| 5 | 1.25 | 125 | 30.33 |

As in the case of ultrafiltration of milk the permeate flux is also decreasing in the diafiltration process during the diafiltration of retentate, when three samples were also taken in every stage. In this section the experimental design showed that the bigger flux (28.34 $l/m^2$*h) is reached when the pressure was 4 bar and the flow rate was 200 l/h (Table 2.).

Table 2. Experimental Design of Nanofiltration of UF retentate (5nm NF/DF)

| Sample | Transmembrane pressure (bar) | Recirculation flow-rate (l/h) | Flux (l/m2*h) |
|---|---|---|---|
| 1 | 2 | 100 | 7.87 |
| 2 | 2 | 200 | 11.98 |
| 3 | 4 | 100 | 20.4 |
| 4 | 4 | 200 | 28.34 |
| 5 | 3 | 150 | 22.11 |

By the comparison of the two diafiltration stages, the flux was similar in both cases. In the diafiltration of the UF permeate the biggest flux was 34.81 $l/m^2$*h as it is illustrated in Table 3. The optimal operation parameters (transmembrane pressure, recirculation flow-rate) are the same in this stage as it was in case of the retentate diafiltration.

Table 3. Experimental Design (5 kDa NF/DF)

| Sample | Transmembrane pressure (bar) | Recirculation flow-rate (l/h) | Flux (l/m2*h) |
|---|---|---|---|
| 1 | 2 | 100 | 21.32 |
| 2 | 2 | 200 | 19.16 |
| 3 | 4 | 100 | 33.58 |
| 4 | 4 | 200 | 34.81 |
| 5 | 3 | 150 | 26.06 |

*3,2 Statistical method*

The results of statistical analysis has shown important information about the correlation between the parameters and reveal any other effect. In the ultrafiltration of milk an effect estimation table was prepared, which are showing the effects of the operation parameters, standard error and p value. The most important parameter is the p value because it shows the transmembrane pressure and the recirculation flow-rate effect on the flux.

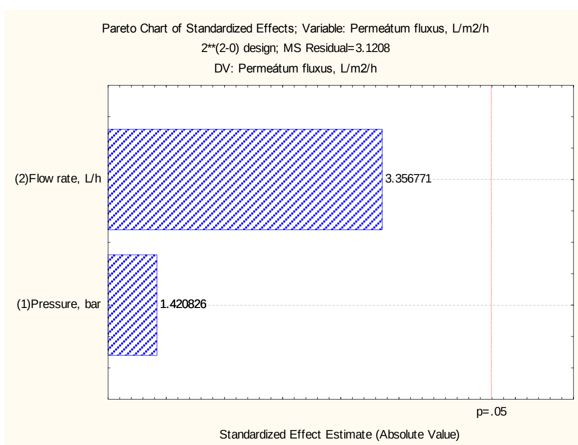


Fig. 1. Pareto chart (UF 100nm)

From the experimental results using Pareto chart were concluded that the recirculation flow rate and transmembrane pressure does not had significant effect on permeate flux, the red vertical line represents the 95% confidence level for the value of p = 0.05.

If the level goes beyond the red line it represent that the effect is significant (Fig.1), which was not true related to both operation parameters (recirculation flowrate and transmembrane pressure), but from the results of the experiments it is obvious that we have to calculate with the effect of recirculation flow rate.

In the second stage of the partial demineralization process, the statistical evaluation was created for the diafiltration of UF retentate. As in the case of ultrafiltration of milk the p value is also bigger than the significant line, even it is very close to p value. By the comparison of the two operation parameters effects, the transmembrane pressure had already significant influence on permeate flux, while the recirculation flow-rate does not had significant effect on the permeate flux as it is illustrated in Fig. 2. It can be seen that the transmembrane pressure have the bigger effect and it is approaching the 95% significance level.

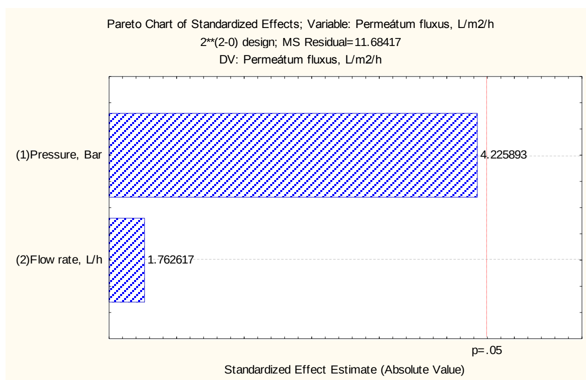


Fig. 2. Pareto chart (Diafiltration 5nm)

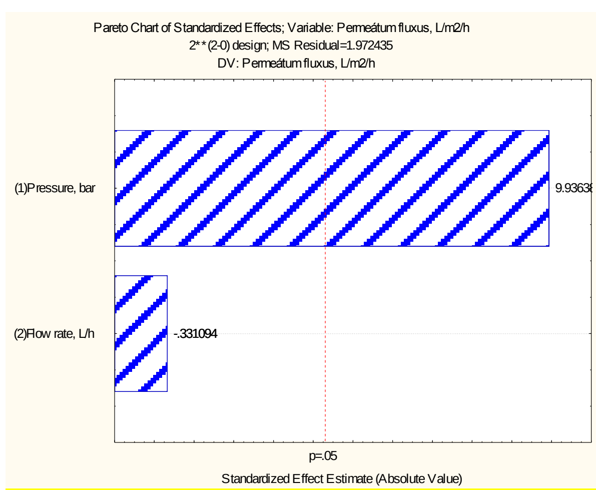


Fig. 3. Pareto chart (Diafiltration 5kDa)

In the diafiltration of permeate the statistical evaluation reveal that the transmembrane pressure has a significant effect on the flux because the p value is smaller than 0.05 as it is illustrated in the Pareto chart (Fig. 3.). The recirculation flow-rate don't have significant effect on the permeate flux.

*4. Conclusions*

From the results of the experiments it can be concluded that the ultrafiltration and also the concentration of the ultrafiltration permeates and retentates by nanofiltration can be successfully achieved by the investigated membranes (100 nm UF, 5nm NF, 5 kDa NF). The examination of the $2^P$experimental designed the statistical evaluation shows that the transmembrane pressure had significant effect on the permeate flux in case of the nanofiltration membranes, while in case of ultrafiltration the recirculation flowrate effected the permeate flux. On the basis of the results of the experiments and the developed simple models can be used for feasibility studies and cost estimation of the cow's milk partial demineralization by using combination of ultrafiltration and nanofiltration.

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