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# Kinetic Modeling of Permeates Flux and Total Hydraulic Resistance of Camel Milk Diafiltration: Effect of pH and NaCl Concentration

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# Abstract

In this study, kinetic modeling of permeates flux and total hydraulic resistance of camel milk diafiltration in different conditions of pH (5.8, 6.3 and 6.8) and NaCl concentration (0, 75 and 150 mM) was performed by using 6 kinetic models. Finally, the homographic kinetic model has been selected for modeling permeates flux and exponential kinetic model has been selected for modeling total hydraulic resistance considering coefficient of determination ( $\mathbb{R}^2$ ) and Root mean square deviation (RMSE). The results of ANOVA of homographic kinetic model illustrated that the linear effect of pH on all model parameters (initial flux  $(J_0)$ , steady-state flux  $(J_{\infty})$ , flux decline time constant (1/b) and flux decline extent (a)) and the linear effect of NaCl concentration at a 95% level on J<sub>0</sub>, 1/b and the interaction effects of pH-NaCl concentration at a 99% level on a were significant. The results of ANOVA of exponential kinetic model also showed that the linear effect of pH had a significant effect on steady-state hydraulic resistance  $(\mathbf{R}_{\infty})$  and the linear effect of NaCl concentration on initial hydraulic resistance  $(\mathbf{R}_{0})$ ,  $\mathbf{R}^{\infty}$  and resistance increment rate (K) and the interaction effects of pH-NaCl concentration at a 95% level on k were significant. According to the results of sensitivity analysis for each 0.1 increase in pH about 1%  $R_{\alpha}$  decreased and for each 10 mM increase in NaCl concentration 2.75%  $R_0$  and 9% 1/b of the samples increased.

Keywords: Camel milk, Diafiltration, Flux, Hydraulic resistance, Kinetic modeling

## Introduction

The use of condensed milk by ultrafiltration in the production of various dairy products, including MPC, requires the consideration of membrane efficiency and changes in its chemical composition during this process (Cheryan, 1998). In some papers, various experimental kinetic models including experimental kinetic models, N-order kinetics and homographic kinetics have been presented to investigate the flux behavior with time and homographic kinetic models, exponential-linear kinetics and exponential kinetics to investigate the behavior of total hydraulic resistance with time (Rajca, Bodzek, & Konieczny, 2009; Razavi, Alghooneh, & Behrouzian, 2017; 2018) that, in the case of the agreement of the experimental data with the proposed models, a better understanding of the flux dynamic behavior and total hydraulic resistance with time in different process conditions can be achieved. Since the efficiency of the ultrafiltration

process is affected by feed properties such as pH and salt concentration and the total hydraulic flux and resistance can be controlled by these factors, therefore, in this study, modeling of flux kinetics and total hydraulic resistance of the diafiltration of camel milk in different pH and salt concentration conditions was performed by 6 kinetic models. Finally, the models with the highest value of  $R^2$  and the lowest value of RMSE were selected and their parameters were discussed in the analysis of the efficiency of the diafiltration process.

## Materials and methods

### Membrane system and operation

In this study, a pilot plant crossflow ultrafiltration system was used. A UF membrane (Model 3838 HFK-131, Koch membrane systems, Inc., USA) made of polysulfone amid (PSA) with MWCO of 20 kDa was applied. The ultrafiltration system was equipped with a feed tank, a centrifugal pump, a flowmeter, a spiral ultrafiltration module, a tubular heat exchanger, two pressure gauges, two flow valves, a digital thermometer, and a digital balance connectable to a PC and printer Camel milk was purchased from a local market in Mashhad, Iran and for camel skim milk production, its fat was separated by a pilot plant milk fat separator in the Food Research Complex, Ferdowsi University of Mashhad. The processes of ultrafiltration and diafiltration of the samples were performed at a transmembrane pressure (TMP) of 80 kPa and a temperature of 20 °C. The diafiltration process was also performed after dilution of the camel milk ultrafiltration stage) and the adjustment of its pH with 0.1 normal lactic acid solution at three levels (5.8, 3.6, and 6.8) and ionic strength by adding sodium chloride (NaCl) at three levels (0, 75, and 150 mM).

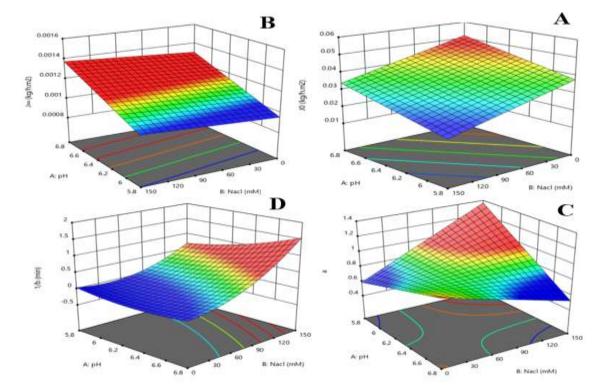
### Experimental design and statistical analysis

In this study, the process treatments were performed in the form of a central composite design (CCD) (5 replications at the central point) for two independent variables at three levels, so that the total number of 13 treatments was obtained (Montgomery, 2001).

#### **Results and discussion**

According to the criteria for selecting the best model, in all samples for different pH and salt conditions, the homographic kinetic model with an  $R^2$  value above 0.90 had the best fitness with the flux-time experimental data. Fig. (1) shows the effect of pH and salt concentration on the parameters of the homographic kinetic model, including initial flux, steady-state flux, flux decline time constant and flux decline extent for different conditions of camel milk diafiltration process.

As shown in Fig. (1), with increasing pH, the parameters  $J_0$  and  $J_\infty$  of the samples increase but the 1/b and parameters of the samples are reduced. Also, according to Fig. (1), with increasing salt concentration,  $J_0$ ,  $J_\infty$  and 1/b of the samples decrease, while a of the samples increases. The results of sensitivity analysis of the obtained models also showed that for every 0.1 increase in pH,  $J_0$  increased by about 2.5% and  $J_\infty$  of the samples increased by 3.1%, and about 13.3% of 1/b and 5.14% of the decrease in flux of samples reduced. Also, according to the results of sensitivity analysis, for every 10 mM increase in salt concentration, about 7.9% of  $J_0$ , 0.46% of  $J_\infty$  and 19.7% of 1/b of the samples decreased and a of the samples increased by about 5.7%. So as the pH increases, due to the increase in the electric potential of the membrane surface, the charges on the surface of the cavities repel each other, resulting in wider cavities and increased J0 and  $J_\infty$  (Kim, Lee, Cho, & Park, 2002).

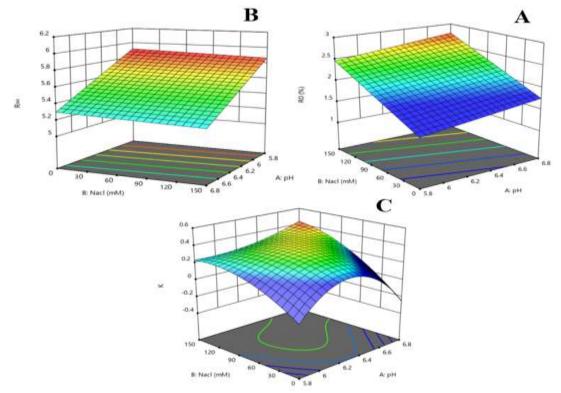


**Fig. 1.** the effect of pH and salt concentration on the parameters of the homographic kinetic model, A: initial flux, B: steady-state flux, C: flux decline time constant and D: flux decline extent for different conditions of camel milk diafiltration process

#### **Total hydraulic resistance**

3 homographic, exponential-linear and exponential models kinetics were also evaluated to investigate the kinetics of hydraulic resistance behavior of all samples. At all levels of pH and salt concentration, the exponential kinetic model with a value of  $R^2$  above 0.90 and the lowest root mean square error (RMSE) had the best fit with the experimental data. Fig. (2) also shows the effect of pH and salt concentration on the parameters of the exponential kinetic model a: initial resistance, b: steady-state resistance and c: the resistance increment rate in the camel milk diafiltration process.

As shown in Fig. (2),  $R_{\infty}$  of the samples increase with decreasing pH however, decreased pH at high salt concentrations reduces the k of the samples. Also, according to Fig. (2),  $R_0$  and 1/b of the samples increase with increasing salt concentration. The results of sensitivity analysis of the obtained models also showed that for every 0.1increase of pH, about 1% of the  $R_{\infty}$  of the samples decreased. Also, according to the results of sensitivity analysis, for every 10 mM increase in salt concentration, about 2.75% was added to  $R_0$  of the samples and k of the samples increases by 9%. Decreased pH reduces the surface charge of particles in low-fat milk such as whey proteins and casein micelles; thus, the repulsive electrostatic forces between proteins and between proteins and the membrane surface are reduced. As a result, proteins tend to settle further on the membrane surface, leading to higher total hydraulic resistance. In addition, the concentration of ionic calcium increases with decreasing pH, therefore, it may increase sediment (Bacchin, Aimar, & Sanchez, 1995).



**Fig. 2.** the effect of pH and salt concentration on the parameters of the exponential kinetic model A: initial resistance, B: steady-state resistance and C: the resistance increment rate in the camel milk diafiltration process

## Conclusions

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The results showed that both homographic and exponential kinetics models had high practical implications in determining process efficiency in all studied conditions and were directly related to the shape characteristics of flux and hydraulic resistance curves and this led to a simple comparison between the shape characteristics of the curves at different pH and salt concentrations. Also, in general, the research results showed that with increasing pH, the values of  $J_0$  and  $J_{\infty}$  of the samples increase, but the parameters  $J_{\infty}$ , 1/b and a of the samples decrease. As the salt concentration increases, the values of  $J_0$ ,  $J_{\infty}$  and 1/b parameters of the samples decrease while  $R_0$ , k and a of the samples increase. Comparing these results with similar studies conducted on cow milk, it can be generally concluded that although camel milk differs greatly from cow milk in terms of physicochemical properties, the overall trends in the dynamic behavior of permeate flux and fouling resistances in the diafiltration process are similar to that of cow milk. Therefore, processing of camel milk by ultrafiltration/ diafiltration process will provide a good market for producers, provided that the process is efficient and cost-effective.

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