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STEADY STATE TRANSMEMBRANE FLUX OF MACROMOLECULAR SOLUTION WITH MICRO AND ULTRA-FILTRATION CONDITIONS

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Abstract. *This study presents results of a micro and ultra-filtration experiment using the food additive known as Guar gum in water solution of 1000 ppm. The transmembrane flux was determined by a laboratory unit which was equipped with a ceramic tubular having a molecular weight cut-off of 500.000 daltons. Experiments were conducted using an average transmembrane pressure of 300 kPa, 400 kPa and 500 kPa for tangential flow rates of 2.4; 3.5 and 5.0 m/s in the turbulent regime flow. Experimental data for fouling (gel layer) and rejection coefficient are presented, and the effects from pressure changes in the process optimization. The transmembrane flux reaches values up to 40% larger in the range of flows studied. Finally, the values of electric condutivity of the permeate flux are indicating significant changes of total ion concentration in permeate and concentrate solution.*

Keywords: *Microfiltration; Ultrafiltration; Guar gum; Mass tranfer*

1. INTRODUCTION

The application of membranes into the separation of biological macromolecules by micro and ultra-filtration is of great importance in bio-products and process industries (Decloux *et al*., 1996). Applications include the concentration of whey proteins for cattle feed and juice fruits (Zeman & Zydney, 1996). Cross flow membrane filtration is a pressure driven separation process where the suspension is circulated parallel to the membrane surface. Such process is commonly classified as micro-filtration, ultra-filtration, nano-filtration or reverse osmose, depending on the size of the solute to be separated. Other conditions applied to the classification of cross flow process are related to the transmembrane pressure.

For practical applications of micro and ultra-filtration, the estimation of membrane fluxes under operational conditions is very important. The accumulation of solute or particles on the membrane surface leads to an effect known as polarization (Zydney, 1997 and Guell & Davis, 1996). Concentration polarization causes a decrease in the hydraulic flux when compared with a flux of a pure solvent alone. This observation has been ascribed to either a reduction in the effective thermodynamic driving force because of the increased osmotic pressure difference

across the membrane, or to an increase in total hydraulic resistance due to the formation of a less permeable phase (gel layer) in series with the membrane (Bader & Veenstra, 1996).

One common feature of these separation process is the existence of a steady state, which is achieved when the concentration polarization reaches its equilibrium condition. This occurs when the flux of solute driven towards the membrane by convection is compensated by back-transport away from the membrane.

Several results have been presented for describing this back flux of solute from the membrane to the bulk suspension (Tanaka, 1997). In the case of micro and ultra-filtration, the relevant transport mechanism away from the membrane is the gradient diffusion. The gradient diffusion depends on both the specific interactions (electrical forces) between particles and the hydrodynamic interactions transmitted by the fluid. This mechanism is specific for process material. In this work we investigated the concentration process in micro and ultra-filtration of Guar gum in aqueous solution, important commercial addictive food (Whistler & BeMiller, 1993). The results are presented for the transmembrane flux in function of pressure transmembrane and Reynolds, and viscosity of concentrate solution and filtrate.

2. EXPERIMENTAL APPARATUS

Figure 1 shows the schematic drawing of the experimental set up for studying hydrodynamics mechanisms in cross-flow. The experimental unit, manufactured by Netzsch Ltda, has two modules containing a 100 cm long and 7 mm diameter tubular ceramic, with membrane surface area 0.022 m^2 (indicated in the Figure by "4"). The fluid of tank (1) was circulated using a positive displacement pump (2). The retentate leaving the module is returned to the feed tank, while permeate rates were measured volumetrically and stored for analysis of the viscosity, electrical conductivity and pH. Therefore, the retentate concentration increases along the time.

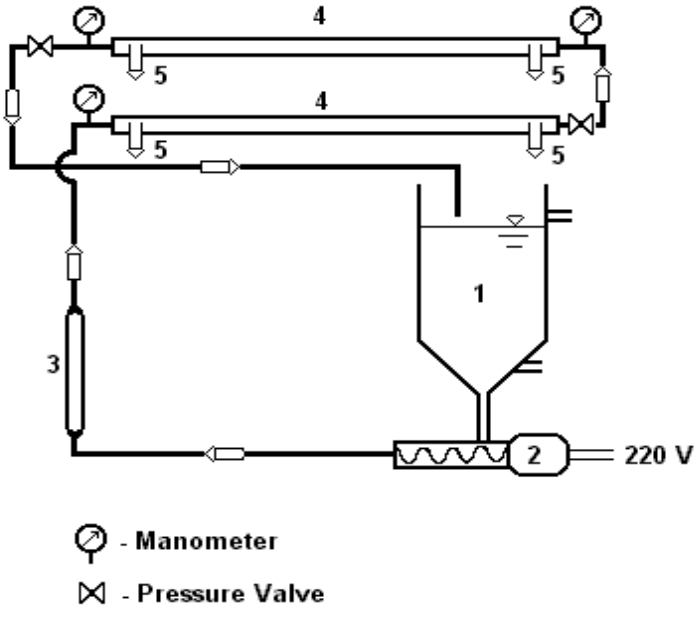


Figure 1. Schematic picture of the experimental apparatus. (1) - tank of fluid; (2) - pump; (3) flowmeter; (4) - membrane module; (5) - exit of the permeate.

The flow rate through the filtration module was monitored by a rotameter (3). The range of medium velocity covered for Guar solution (1000 ppm) is presented in turbulence regime flow. Two manometers were used to measure inlet and outlet pressures, the transmembrane pressure (ΔP_{tm}), is the mean of inlet and outlet pressures.

The measurements were performed at 30 ± 0.8 ^{o}C, and using only one membrane module (surface membrane area, 0.011 m^2). The nominal molecular weight cut-off is $5x10^5$ daltons (0,6 μm respectivity), and the membrane water permeability 120 l/h at 400 kPa and 3.5 m/s (mean velocity in tube).

Experiments were carried out with Guar gum macromolecular solution at 1000 ppm, of average molecular weight 10^6 daltons. The viscosity measurements were made using a rotational cylindrical concentric, model LVDII+ (Spindle SC18) Brookfield. A clear permeate flow into the shell side of the membrane module was obtained, so that permeate viscosity equalled the water viscosity, 1 cPoise (10^{-3} Pa.s).

Between experiments, the membranes were regenerated by the following procedure: i-) 40 minutes in flow washing with neutral detergent solution at 2% of concentration; ii-) 12 hours in static washing with enzymatic detergent at 2% of concentration.

3. RESULTS AND DISCUSSION

Figure 2 presents the results of accumulated filtrate flux (l/h.m^2) in function of time (min) for the Guar gum solution (1000 ppm). The results of the filtration process were accomplished in nine different conditions, for three rate flux and transmembrane pressures values. The values are different in the graphical from the rate flux of the flow in tube (different shear stress), and the corresponding value of the transmembrane pressure. It is observed for the corresponding curves of the rate of flow of 5.0 m/s, that the variation rate is maintained practically constant. On the other hand, we have observed a decline in the flow rate, after 20 minutes of the beginning of the process, identified in picture as a non lineal variation.

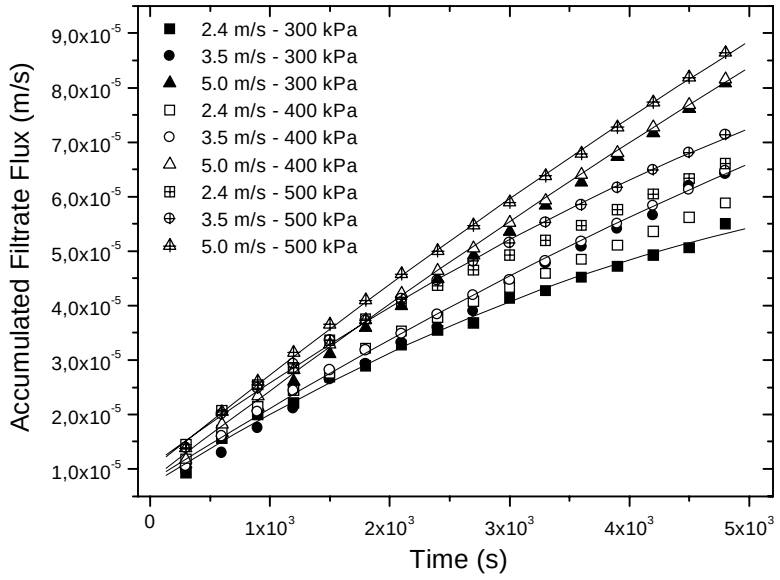


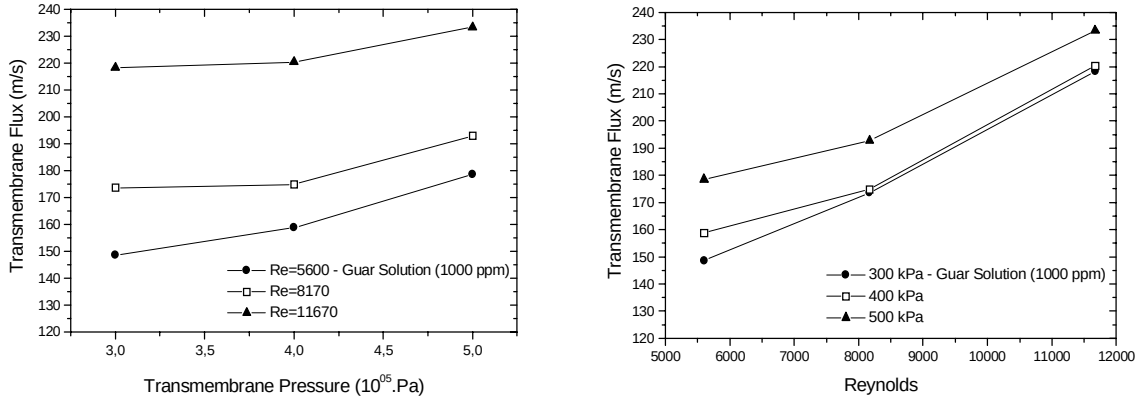
Figure 2. Graphical of Accumulated Filtrate Flux (m/s) in function of Time (s) for the Guar gum solution (1000 ppm).

The results demonstrate that the effect of the shear stress in the membrane surface is satisfactory in the process of concentration of the product, provoking increments in the permeate flow, and avoiding the formation of the polarized layer of resistance gel. Starting from the increase of the transmembrane pressure, a satisfactory increase is observed in the final value of the final permeate flow.

Experimental results are valid in the micro-filtration process (300 kPa) and ultra-filtration (400 kPa and 500 kPa), specific for the turbulent regime flow. In typical conditions or ultra-filtration, the best filtration process is obtained for Re=11870 and ΔP_{tm} =500 kPa. The corresponding fitting is given by: $J = 1.18 \cdot 10^{-05} + 1.57 \cdot 10^{-08} \cdot t$.

Figure 3 shows results of transmembrane flux (m/s) as functions of transmembrane pressure (kPa) (a), and Reynolds number (b) for Guar gum solution in 1000 ppm. In the Fig. 3.a and 3.b it is observed that the transmembrane flow increases with the increase of the Reynolds number, up to about 50%.

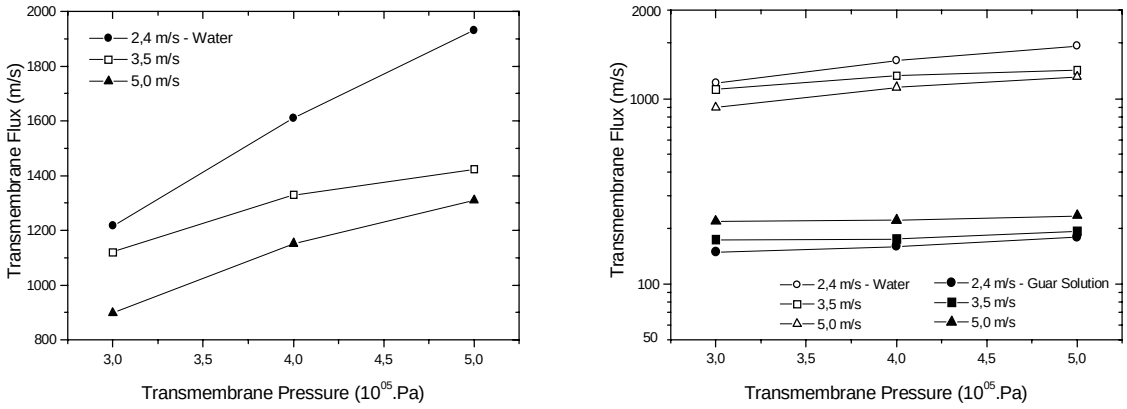
The Fig. 3.b indicates that there is no variation in the flow due to the increase of the transmembrane pressure from 300 to 400 kPa. The increase of the transmembrane pressure to 500 kPa provokes increase in the transmembrane flow. These effects are related with the formation and resistance of the gel-polarized layer in the membrane surface. The model most commonly used in the literature relates the transmembrane flux (J) in function of the transmembrane pressure (ΔP_{tm}) in the form: $J = (\Delta P_{tm})/(R_h+R_g)$; where R_h is the hydraulic resistance and R_g is the resistance to the flow due to gel-polarized layer.



(a) (b)

Figure 3. (a) Graphical of Transmembrane Flux (m/s) in function of Transmembrane Pressure (10^{05}.Pa); (b) Graphical of Transmembrane Flux (m/s) in function of Reynolds number. Guar gum solution (1000 ppm).

In Fig. 4 the results of the transmembrane flux (m/s) as function of transmembrane pressure (kPa) for water flow are presented. In Fig. 4.b a comparison is made with the results of the Guar gum solution. The values of transmembrane flux of pure water are higher at about 400%. The results of the hydraulics resistance are shown in the Table 1.



(a) (b)

Figure 4. (a) Graphical of Transmembrane Flux (m/s) in function of Transmembrane Pressure (Pa) for Water; (b) Graphical of Transmembrane Flux (m/s) in function of Transmembrane Pressure (Pa) for Guar gum solution (1000 ppm).

In Table 1, the values of R_h (water) and R_g (Guar gum solution - 1000 ppm) are presented. The R_g value for Re=11670 is approximately 100% inferior to the corresponding value for Re=5600, indicating the satisfactory action of the shear stress in decreasing the effect of the polarized layer gel. The values of R_h and R_g were obtained by linear fitting of the data of Fig. 4, having correlation coefficients larger than 0.95.

Table 1. Values of hydraulic and gel layer resistances.

Re*	Water (R_h)	Guar gum solution 1000 ppm (R_g)	Relative increment in the global hydraulic resistence (R_h+R_g) (%)
5600	357.5	14.9	4.2
8170	151.4	9.6	6.3
11670	39.4	7.5	19.0

* The value of Re corresponds to the gum Guar solution.

Dimensional analysis show that two dimensionless quantities ($\Delta P_{tm} / \rho u^2$) and ($R_h \cdot J / \Delta P_{tm}$) can be derived for the studied phenomena. The first quantity is similar to the inverse of the Euler number, sometimes called the "number of units of energy". ρu^2 is the minimum energy per unit volume required to transport the fluid through the tubular membrane at the velocity "u", while "ΔP_{tm}" is the energy per unit volume dissipated in the water transport through membrane. $\rho u^2 / \Delta P_{tm}$ compares the shear stress to the driving pressure; it will then be called the shear stress number. The quantity $R_h \cdot J / \Delta P_{tm}$ will be called the resistance number (Elmaleh, et alli, 1998), and will be written in this work as J / u.

In Fig. 5 the results are presented of the resistance number (J / u) as function of the inverse of shear stress number ($\Delta P_{tm} / \rho u^2$). The results were obtained for the accumulated transmembrane flow for the initial twenty minutes of the process, and for whole the process, corresponding to eighty minutes. The results indicate that resistance number depends on the inverse of shear stress number as a linear decay function, considering the vertical coordinate

in logarithmic scale. The graphical presented in Fig. 5 has shown to be of great importance due to the distinction of behaviors among aqueous solutions and membrane systems (Elmaleh, et alli, 1998). In this work a positive slope between (J/u) and ($\Delta P_{tm}/\rho u^2$) can indicate a complete elimination of the polarized layer, probably because of the satisfactory effect of the turbulent shear stress in the membrane surface.

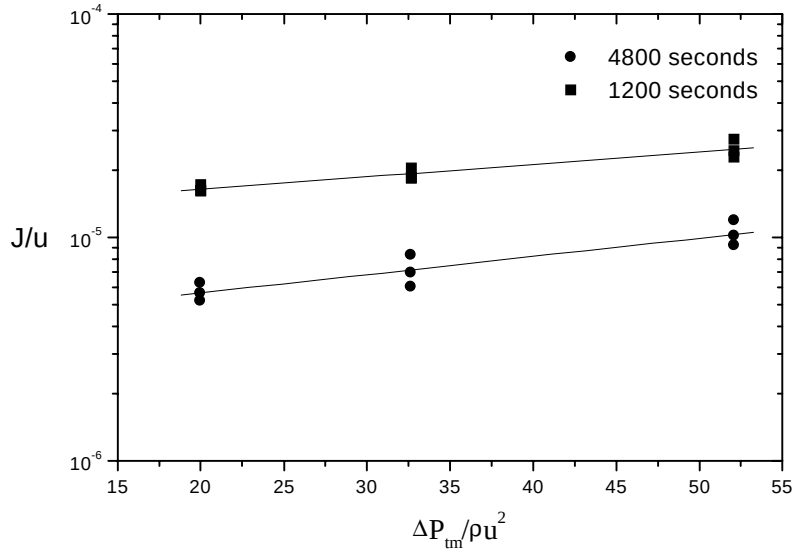


Figure 5. Graphical of resistance number in function of the inverse of shear stress number.

The correlation equations (apparent linear fitting) for the two cases are: $\log(J/u) = -5.41 + 8.18 \cdot 10^{-03} \cdot (\Delta P_{tm}/\rho u^2)$ (1200 seconds) and $\log(J/u) = -4.90 + 5.56 \cdot 10^{-03} \cdot (\Delta P_{tm}/\rho u^2)$ (4800 seconds), having correlation coefficients larger than 0.92 for the two cases.

The mechanism of concentration in macromolecular solution was also investigated studying the viscosity of the permeate, and the concentrate from the beginning of the process. The viscosity is an important physical parameter in the crossflow filtration process (Ilias, *et al*., 1995 and Charcosset & Choplin, 1996). In Figs. 6.a, 6.b and 6.c the values of the viscosity of the permeate, concentrate and aqueous solution of gum Guar (1000 ppm) are presented. The results are presented for three processes accomplished in different rate fluxes, or Re=5600 (2.4 m/s), Re=8170 (3.5 m/s) and Re=11670 (5 m/s).

In Fig. 6.d the values are the average of all the values presented in 6.a, 6.b and 6.c. The filtrate or permeate presents values of viscosity close to the water, while the concentrate solution values about 35% larger than of the initial aqueous solution (1000 ppm).

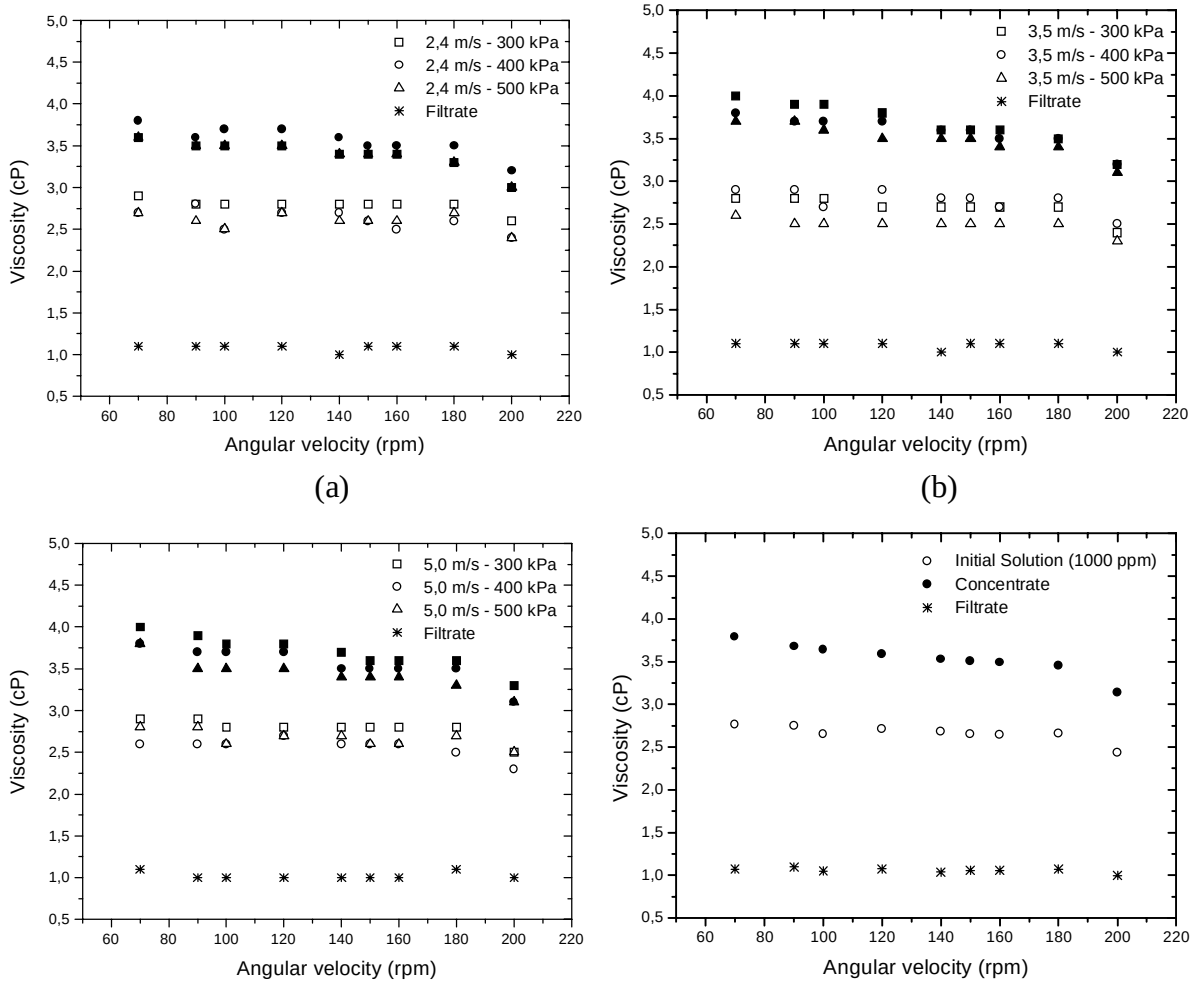


Figure 6. Graphical of Viscosity (cP) in function of angular velocity (rpm). (a) Re=5600; (b) Re=8170; (c) Re=11670; (d) Mean values of all presented in (a), (b) and (c).

The operating temperature was maintained at 30 ± 0.8 ^{o}C, with the aid of the re-circulation of cold water through the mantle tank. In Fig. 7 the shear stress results (N/m^2) are plotted in function of shear rate (1/s) corresponding to the values presented in the Fig. 6.d.

The literature (Whistler & BeMiller, 1993) classifies solutions of Guar gum as newtonian. The result presented in Fig. 7 (of initial shear stress smaller than 0.09 N/m^2), is probably related to the fact that Guar gum solutions are newtonian fluids but additional experiments are necessary in order to confirm our observations.

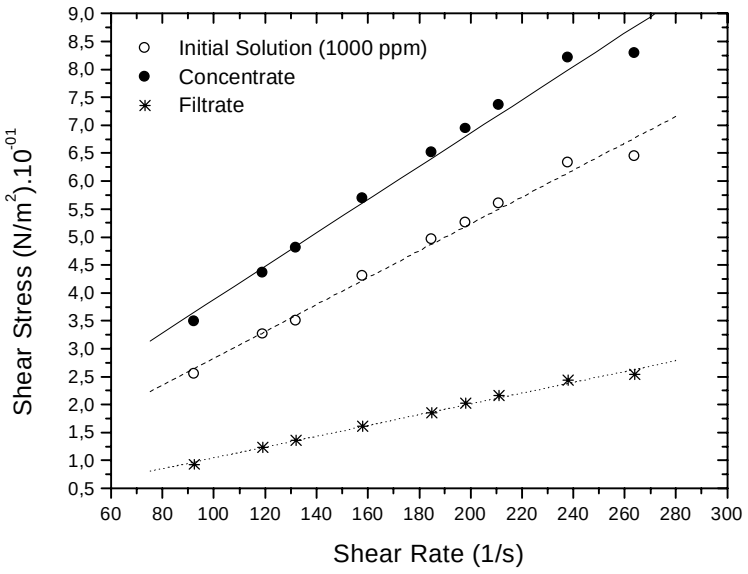


Figure 7. Graphical of Shear Stress (N/m^2) in function of Shear Rate (1/s) for the average values of the three process presented in Figure 6.d.

In Table 2 the values of the electric conductivity (10^{-06} Siemens) are presented for all the experiments. The results refer the average of three values accomplished at the end of the process with Hanna electrical conductivity (HI-8733). It is observed that the values of conductivity of the concentrated solution are larger than the ones of the filtrate, indicating the largest concentration of ions in the concentrate solution. PH values were also accompanied in each experiment They, the pH values in the end of the process. The results didn't demonstrate significant differences between the concentrated solution and the filtrate, and a mean value of the concentration solution was of pH=8.17.

Table 2. Values of electrical conductivity for permeate and concentrate solution.

Experiments	Filtrate or Permeate (μS)	Concentrate Solution (μS)
1	228	240
2	230	238
3	229	248
4	230	243
5	234	245
6	226	239
7	225	241
8	234	244
9	234	241
Mean Value	230	242.1

4. CONCLUSION

We have presented the experimental data and the analysis of cross-flow filtration of Guar gum macromolecular solution (1000 ppm). The results indicate that transmembrane flux depends on medium flux or Reynolds number in all the studied cases. The best filtration process is obtained for Re=11870 and ΔP_{tm} =500 kPa (typical conditions for ultra-filtration). The fitting for the best process condition is given by: $J = 1.18 \cdot 10^{-05} + 1.57 \cdot 10^{-08} \cdot t$ (Re=11870 and 500 kPa) and $J = 9.47 \cdot 10^{-06} + 1.51 \cdot 10^{-08} \cdot t$ (Re=11870 and 400 kPa). In filtration process, we have quantified the increase in viscosity as being 35 % in the initial solution. The hydraulics and gel layer resistances were presented, and the smallest value for R_g was obtained at Re=11879. Therefore, the increase of shear stress is satisfactory for the reduction of the gel layer. This result was also confirmed by the analysis of the dimensionless groups presented in the Fig. 5. Electrical conductivity and pH were measured, for the concentrate solution and the permeate. The conductivity values of concentrated solution are approximately 11% larger than the filtrated solution, while the pH stayed practically constant in the value of 8.17.

Acknowledgement

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