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Separation Science and Technology
Publication details, including instructions for authors and subscription information:
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Experimental Data and Behavior of Starch/Water Solutions with an Ultrafiltration Module

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To cite this Article Gomez-Gotor, A. , Susial, P. , Guerra, A. and Ibarra, B.(1996) 'Experimental Data and Behavior of Starch/Water Solutions with an Ultrafiltration Module', Separation Science and Technology, 31: 2, 173 — 188
To link to this Article: DOI: 10.1080/01496399608000688
URL: http://dx.doi.org/10.1080/01496399608000688

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SEPARATION SCIENCE AND TECHNOLOGY, 31(2), pp. 173–188, 1996

Experimental Data and Behavior of Starch/Water Solutions with an Ultrafiltration Module

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ABSTRACT

A crossflow, plane membrane, ultrafiltration module is presented in this paper. The experimental data obtained for different concentrations of the starch/water system after modifying the transmembrane pressure and the feed inlet velocity are given. A series resistances model was applied to the experimental data in order to obtain analytically the contributions of the fouling and gel resistances. In addition, the experimental data were correlated to a diffusion model. By considering the phenomenon of concentration-polarization, this allowed us to evaluate the concentration of the gel layer to each composition of the feed flow and the mass transfer coefficients, as well as their relation with the feed inlet velocity. The density and viscosity of various solutions of the starch/water system were determined. This enabled us to evaluate the diffusion coefficient after using the Grober and Chilton–Colburn equations.

Key Words. Membrane processes; Ultrafiltration; Diffusion; Mass transfer

INTRODUCTION

One of the probable reasons for the advantageous competition of membrane processes (MP) with other substance separation techniques is based

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on the fact that with MP it is not necessary to modify the phases of the substances which are put in contact, thus reducing the energetic costs. Also, substances separated by MP are neither degraded or deteriorated, and these separation processes are nonpolluting. These advantages encourage their study and development.

At present, substance separation techniques which use membranes are being recognized for their importance by industry. Some applications of MP can be found in the food industries (milk concentration for cheese production, concentration of fruit juices, purification of drinks, etc.), as well as in medicine and pharmacy (to obtain blood plasma, enzyme concentration, and the purification, production, and concentration of vaccines and antibiotics, etc.), in the automation and electronic industries, and for the treatment of water (purification, desalination, and/or reuse).

Ultrafiltration (UF) is one of the substance separation techniques used in MP. It is conceptually based on the inversion of the natural and spontaneous process of substance redistribution between nonmixeable phases when a driving force exists between them. The inversion of the spontaneous process is possible if a force greater than the driving gradient is applied to the system.

Non or partial mixability of solutions brought in contact in UF is achieved by utilizing a membrane (a physical device) of different materials, semipermeable to the solutions, and acting as a barrier that fractionates the solution components according to their size and shape (1).

Thus, UF as unit operation can be characterized by the applied external force (200–1000 kPa, approximately) or by the approximate solute size that the physical device can retain (10^3–10^6 Dalton) (2).

An experimental UF installation which utilizes plane membranes is described in this work. We also adduce experimental data of starch/water solutions. These solutions, in the range between 42.5 and 850.0 ppm, were recirculated in crossflow filtration while modifying the feed inlet velocity between 0.99 and 1.74 m/s at a constant temperature of 298 K. The pressure of the incoming flow was also modified from 200 to 500 kPa while maintaining constant temperature, concentration, and velocity conditions of the feed solution.

Experimental data of the starch concentrations in the permeate and the volume flux were obtained for each of the operating conditions within the indicated ranges. This enabled evaluation of the gel layer and fouling layer by using a phenomenological model of resistances (3, 4) as well as the mass-transfer coefficients and the rejected solute capacity of the membrane considering a diffusive model (5, 6). These values could be of interest for the chemical engineer in the design of installations.

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EXPERIMENTAL SECTION

Materials

Pure water obtained via distillation and treated thereafter with an ion exchanger (resistivity higher than 5 MΩ·cm) has been used in the experimental work. Soluble starch (reagent grade, ref. 171096) came from the commercial company Panreac.

Cellulose acetate plane membranes from Dow Danmark A/S, type CA600PP, were used. They have a molecular weight cut-off of 20,000 Dalton and an area of 8.4×10^{-3} m^2.

The manufacturer's recommendations for operation limits and membrane cleaning and disinfection were always taken into account. However, different cleaning cycles were done successively in order to maintain the initial hydropermeability conditions of the membranes.

Solutions with 1000 ppm hydrogen peroxide (Panreac, (puriss., ref. 141076) were used for the disinfection of membranes, kept at 298 K, and recirculated at a pressure of 500 kPa. Pure water, as described above, was used at 298 K and recirculated at 500 kPa during the membrane washing cycles. After each operating period the disinfected and washed membranes were stored in the experimental installation with sodium bisulfite solutions (Panreac, puriss., ref. 141698).

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Equipment and Procedure

A UF module (DDS Minilab-10) with two plane membranes from Dow Danmark A/S and the corresponding pressure gauges were used. The UF module (made of stainless steel and polysulfone), of dimensions 198×10^{-3}, 127×10^{-3}, and 45×10^{-3} m, has an internal volume of 28.5×10^{-6} m^3 and is able to operate at pressures up to 700 kPa.

The DDS module consists of two sections, one on top of the other, connected in series. Each section has four channels of the DDs type with a cross section of 5.37×10^{-5} m^2 and a hydraulic diameter of 2.3×10^{-3} m (average values). Each channel is connected to the feed flow by means of a gap with a diameter of 5×10^{-3} m. The membrane is located above the piping and above the membrane is the separating base.

The feed flow received by the Minilab-10 module is driven by a pump with a motor and velocity selector Eco Gearchem (mod. 64) of 0.22 kW, which is able to reach a maximum pressure of 700 kPa.

The experimental installation, shown in Fig. 1, permits continuous operation of a batch in a closed loop. Rotameters (Gilmont Instruments) were used to measure the permeate and feed flows.

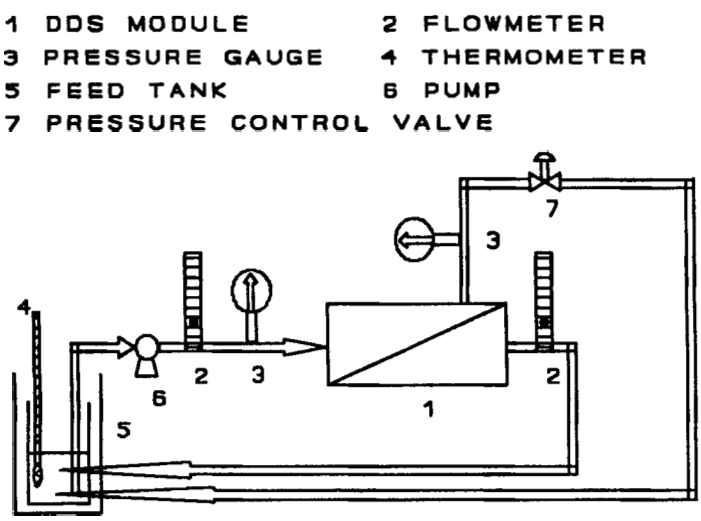

FIG. 1 Flow diagram of UF experimental set-up.

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For the solute concentrations (42.5, 85.0, 170.0, 425.0, and 850.0 ppm) at the feed side, the solute concentrations in the permeate were measured for each of the pressure values (200, 300, 400, and 500 kPa) and the velocities of the feed solution (0.99, 1.24, 1.49, and 1.74 m/s) by a total organic and inorganic carbon analyzer (TOC 700 Analyzer, O. I. Corporation) in which different loops were used in order to make determinations possible in the $C_0 \leq 100$, $C_0 \leq 500$, and $C_0 \leq 1000$ ppm ranges.

Before making the experimental determinations, the steady-state (SS) condition was evaluated. For this purpose, a feed inlet solution with 500 ppm starch was used, thermostated at 298 K, held at a constant operation pressure (200 kPa) and feed inlet velocity (0.99 m/s). After about 3 hours of recirculating the solution across the installation at the indicated conditions, the permeate velocities and concentrations were measured at regular time intervals; the results are shown in Fig. 2. Analysis allowed us to estimate that SS between two consecutive samples is reached after recirculation intervals longer than 20–25 minutes. Thus, in the present study, an interval of 30 minutes was chosen.

The coefficient of hydropermeability of the membrane, $L_p = 1.17 \times 10^{-10}\ Pa^{-1} \cdot m \cdot s^{-1}$, was determined before starting the experimental work with the starch solutions (Fig. 3). Then a solution of analyzed concentration was prepared, the feed inlet velocity was fixed, and the transmembrane pressure was increased with the retentate side valve, starting at the

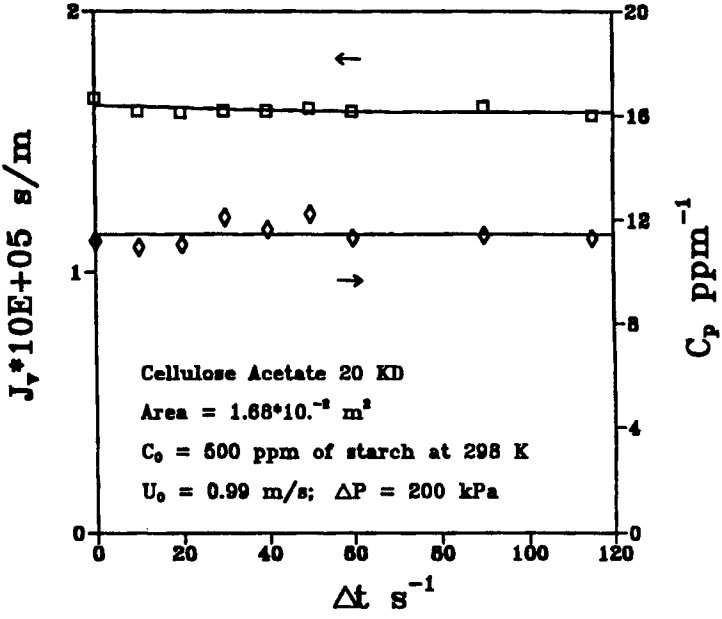

FIG. 2 Experimental data for the evaluation of the steady state.

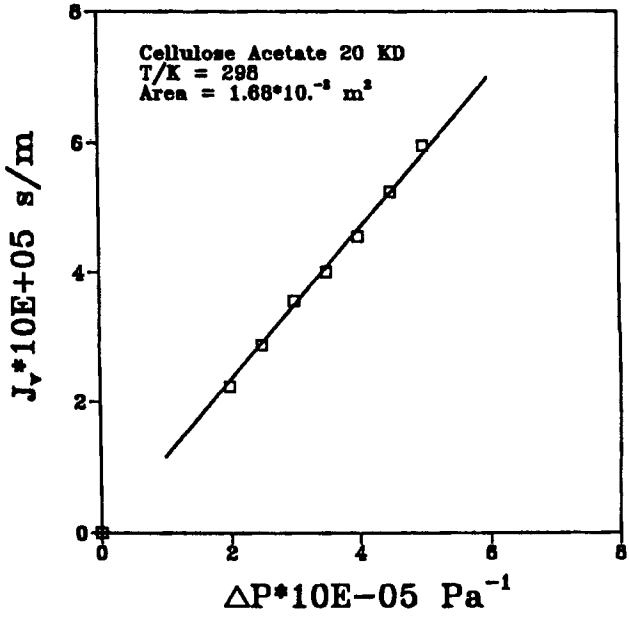

FIG. 3 Determination of the hydraulic permeability coefficient.

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lowest pressure point. For each selected pressure value and after the time interval necessary to reach SS, the permeate flux was determined and its concentration analyzed.

The same procedure was followed for several increasing feed inlet velocities as well as for other feed inlet solute concentrations. In all cases the experiment started with the lowest concentration and ended with the highest feed inlet solute concentration.

The experimental work was carried out during several consecutive days. After finishing each experiment the membranes were washed and disinfected during several cycles and stored in sodium bisulfite (2500 ppm) solutions. Before starting another experimental determination, the membranes were again washed and their coefficients of hydropermeability were determined. If the recovery of the membranes was not higher than 85%, they would have been replaced (7), but this eventuality did not occur.

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RESULTS AND DATA PROCESSING

The experimental data of J_V with C_P of the water–starch solutions with concentrations of 42.5, 85.0, 170.0, 425.0, and 850.0 ppm at the feed inlet side were obtained in a UF module for feed inlet velocities of 0.99, 1.24, 1.49, and 1.74 m/s and pressure values of the incoming flow of 200, 300, 400, and 500 kPa (Table 1), following the procedure described above.

Assuming that solutions of macromolecular solutes generally show low osmotic pressures when compared, in similar weight percentage conditions, with saline solutions of low molecular masses (8), and considering the low inlet solute concentrations used, as well as the molecular mass of starch (between 100 and 300 times the molecular mass of glucose), the values corresponding to the osmotic pressures were not taken into account in the present work.

Phenomenological Model of Resistances

The behavior of the permeate flux considered for each flow velocity presents a quasi-linear evolution with respect to the transmembrane pressure, mainly at very low pressures. The observed linearity is a consequence of the absence of fouling, assuming fouling is exclusively by solute adhering to the membrane surface. However, even at very low values of feed inlet solute concentrations, as in the present study, other aspects have to be considered: total or partial blocking, plugging, or scaling of the interstitial spaces of the polymeric matrix, as well as the impermeability of the concentrated film, its compaction and its polarization, since these are effects that have to be analyzed (9, 10).

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TABLE 1
Experimental Data of the Starch/Water Solutions with the UF Plane Membrane

U_0 = 0.99 m/s			U_0 = 1.24 m/s			U_0 = 1.24 m/s			U_0 = 1.24 m/s		
ΔP (Pa)	J_V ($m^3 \cdot s^{-1}$)	C_P ($kg \cdot m^{-3}$)	ΔP (Pa)	J_V ($m^3 \cdot s^{-1}$)	C_P ($kg \cdot m^{-3}$)	ΔP (Pa)	J_V ($m^3 \cdot s^{-1}$)	C_P ($kg \cdot m^{-3}$)	ΔP (Pa)	J_V ($m^3 \cdot s^{-1}$)	C_P ($kg \cdot m^{-3}$)
					C_0 = 42.5E−03 kg/m³						
4.97E+05	8.24E−07	8.7E−03	4.95E+05	8.74E−07	8.1E−03	4.95E+05	9.15E−07	6.8E−03	4.85E+05	9.49E−07	4.4E−03
3.97E+05	7.01E−07	8.4E−03	3.92E+05	7.08E−07	7.1E−03	3.87E+05	7.16E−07	5.7E−03	3.85E+05	7.35E−07	4.3E−03
2.95E+05	5.15E−07	6.9E−03	2.90E+05	5.21E−07	6.2E−03	2.85E+05	5.24E−07	4.9E−03	2.80E+05	5.32E−07	4.2E−03
1.95E+05	3.37E−07	6.7E−03	1.90E+05	3.42E−07	5.7E−03	1.85E+05	3.45E−07	4.5E−03	1.80E+05	3.48E−07	3.7E−03
					C_0 = 85.0E−03 kg/m³						
4.97E+05	7.53E−07	10.0E−03	4.95E+05	7.91E−07	8.7E−03	4.90E+05	8.19E−07	7.5E−03	4.85E+05	8.52E−07	6.2E−03
3.97E+05	6.04E−07	9.7E−03	3.92E+05	6.38E−07	7.0E−03	3.87E+05	6.65E−07	6.0E−03	3.85E+05	6.85E−07	4.8E−03
2.95E+05	4.79E−07	9.5E−03	2.90E+05	4.90E−07	6.6E−03	2.85E+05	5.02E−07	5.8E−03	2.80E+05	5.20E−07	4.7E−03
1.95E+05	3.10E−07	6.9E−03	1.90E+05	3.20E−07	5.9E−03	1.85E+05	3.27E−07	5.7E−03	1.80E+05	3.32E−07	4.6E−03
					C_0 = 170.0E−03 kg/m³						
4.97E+05	6.80E−07	16.0E−03	4.95E+05	7.11E−07	15.6E−03	4.90E+05	7.50E−07	13.1E−03	4.85E+05	7.71E−07	10.7E−03
3.97E+05	5.61E−07	13.3E−03	3.92E+05	5.85E−07	12.7E−03	3.87E+05	6.04E−07	10.6E−03	3.85E+05	6.36E−07	7.9E−03
2.95E+05	4.52E−07	10.7E−03	2.90E+05	4.57E−07	9.2E−03	2.85E+05	4.66E−07	8.3E−03	2.80E+05	4.77E−07	7.0E−03
1.95E+05	2.86E−07	8.5E−03	1.90E+05	2.92E−07	7.4E−03	1.85E+05	2.99E−07	6.9E−03	1.80E+05	3.05E−07	6.3E−03
					C_0 = 425.0E−03 kg/m³						
4.97E+05	6.24E−07	29.1E−03	4.95E+05	6.55E−07	25.9E−03	4.90E+05	6.83E−07	20.4E−03	4.85E+05	7.11E−07	16.5E−03
3.97E+05	5.23E−07	18.4E−03	3.92E+05	5.45E−07	16.2E−03	3.87E+05	5.68E−07	14.1E−03	3.85E+05	5.93E−07	12.4E−03
2.95E+05	4.22E−07	14.4E−03	2.90E+05	4.29E−07	12.5E−03	2.85E+05	4.36E−07	11.0E−03	2.80E+05	4.44E−07	9.8E−03
1.95E+05	2.72E−07	10.5E−03	1.90E+05	2.78E−07	9.5E−03	1.85E+05	2.85E−07	8.4E−03	1.80E+05	2.91E−07	7.6E−03
					C_0 = 850.0E−03 kg/m³						
4.97E+05	5.66E−07	57.3E−03	4.95E+05	5.85E−07	50.1E−03	4.90E+05	6.04E−07	42.5E−03	4.85E+05	6.30E−07	35.0E−03
3.97E+05	5.03E−07	48.6E−03	3.92E+05	5.13E−07	37.0E−03	3.87E+05	5.22E−07	30.0E−03	3.85E+05	5.28E−07	27.0E−03
2.95E+05	3.90E−07	35.0E−03	2.90E+05	3.98E−07	27.7E−03	2.85E+05	4.03E−07	24.5E−03	2.80E+05	4.09E−07	22.0E−03
1.95E+05	2.61E−07	22.1E−03	1.90E+05	2.63E−07	19.4E−03	1.85E+05	2.65E−07	16.6E−03	1.80E+05	2.68E−07	15.0E−03

A reduction of the permeate flux with respect to time is observed (Fig. 2). This variation increases with increasing pressure and increases even more with an increase of concentration at the feed side. The following phenomenological model, which considers different contributions by means of series resistances, is consistent with the behavior of the fouling and gel layer resistances and their occurrences in the evolution of the permeate flux (3, 4):

$$J_V = \frac{\Delta P}{R_M + R_F + R_G} \tag{1}$$

The fouling resistance has been considered to be due to a solute film adhering to the membrane (11), and originates from the presence of interfacial forces which can include total or partial blocking effects of the interstitial spaces of the membrane polymeric net. This film is assumed to be undeformable and unsensible to the applied pressure, but depends on solute–solute and solute–membrane interactions, and is thus a function of the solute concentration at the feed side and of its circulation velocity according to the expression

$$R_F = \phi_{RF} C_0^a U_0^b \tag{2}$$

The solute concentration decreases in the so-called gel layer between the fouling film and the bulk solution. The solute retained in this region is continuously renovated by the feed inlet recirculation. However, its concentration profile is time-independent after SS is achieved. The structure of the gel layer can be assumed to result from the addition of an infinite number of thin films of constant concentration which increases as they approach the fouling film. Nevertheless, this layer is presented as a single compressible layer which offers a global resistance to the permeate flux, considering a concentrated film behavior (12, 13), which depends on feed inlet solute concentration, feed inlet velocity, and applied pressure (3).

$$R_G = \phi_{RG} C_0^c U_0^d \Delta P \tag{3}$$

In order to correlate all the experimental data simultaneously, taking into account Eqs. (1)–(3) and using the inverse of the coefficient of hydropermeability for the membrane resistance, a multiple nonlinear regression procedure was applied (14) which minimizes the matching error by means of the objective function,

$$F = \sum \left[\left(\frac{\Delta P}{J_V} - R_M \right)_{EXP} - \left(\frac{\Delta P}{J_V} - R_M \right)_{CALC} \right]^2 \tag{4}$$

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resulting in the objective function, $F = 1.09 \times 10^{19}$, and the volume flux,

$$J_V = \frac{\Delta P}{(0.855 \times 10^{10}) + (0.312 \times 10^{10} C_0^{0.323} U_0^{-1.602}) + (0.724 \times 10^4 C_0^{0.606} U_0^{-0.019} \Delta P)} \quad (5)$$

The experimental data and those calculated with Eq. (5) were represented graphically (e.g., Fig. 4). An increasing relationship between the feed inlet velocity and the permeate flux, as well as a decreasing relationship between the permeate flux and the feed solute concentration, can be observed. Evaluation of the contributions of the resistances (Table 2) for each of the pressure values at the inlet shows that the resistance of the gel layer is lower than the fouling resistance, except for high concentrations and pressures. Its value increases with concentration and shows a faster growth with concentration than with fouling resistance.

Diffusion Model

Different models have been proposed to explain MP behavior by applying the diffusion concept (15, 16). The molecular redistribution process

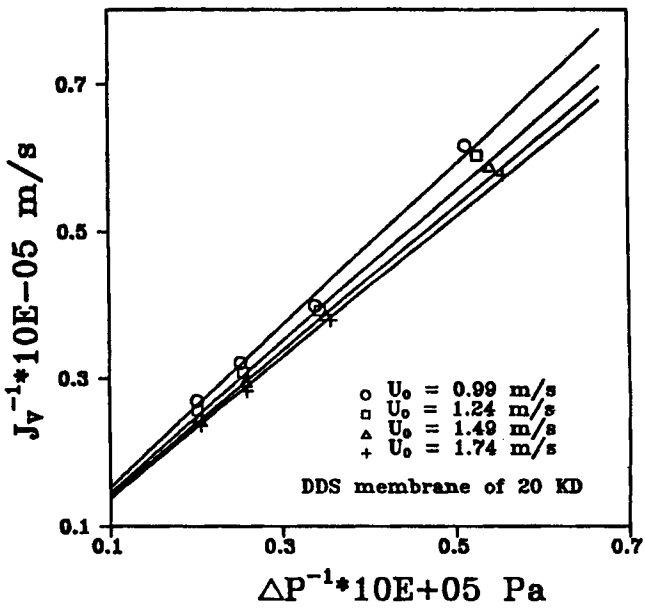

FIG. 4 Correlation of experimental data for 425.0 ppm of starch at 298 K with the series resistances model.

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TABLE 2
Results Obtained for the Fouling and Gel Resistances

U_0 (m·s^{-1})	R_F (Pa·m^{-1}·s)	R_G (Pa·m^{-1}·s) $\Delta P = 2 \times 10^5$ Pa	$\Delta P = 3 \times 10^5$ Pa	$\Delta P = 4 \times 10^5$ Pa	$\Delta P = 5 \times 10^5$ Pa
		C_0 = 42.5E−03 kg/m^3			
0.99	0.114E+10	0.214E+09	0.320E+09	0.427E+09	0.534E+09
1.24	0.797E+09	0.213E+09	0.319E+09	0.425E+09	0.532E+09
1.49	0.594E+09	0.212E+09	0.318E+09	0.424E+09	0.530E+09
1.74	0.463E+09	0.211E+09	0.317E+09	0.423E+09	0.528E+09
		C_0 = 85.0E−03 kg/m^3			
0.99	0.143E+10	0.325E+09	0.488E+09	0.650E+09	0.813E+09
1.24	0.997E+09	0.324E+09	0.486E+09	0.648E+09	0.809E+09
1.49	0.743E+09	0.323E+09	0.484E+09	0.645E+09	0.807E+09
1.74	0.579E+09	0.322E+09	0.483E+09	0.643E+09	0.804E+09
		C_0 = 170.0E−03 kg/m^3			
0.99	0.179E+10	0.495E+09	0.742E+09	0.990E+09	0.124E+10
1.24	0.125E+10	0.493E+09	0.739E+09	0.986E+09	0.123E+10
1.49	0.929E+09	0.491E+09	0.737E+09	0.982E+09	0.123E+10
1.74	0.725E+09	0.490E+09	0.734E+09	0.979E+09	0.122E+10
		C_0 = 425.0E−03 kg/m^3			
0.99	0.241E+10	0.862E+09	0.129E+10	0.172E+10	0.216E+10
1.24	0.168E+10	0.859E+09	0.129E+10	0.172E+10	0.215E+10
1.49	0.125E+10	0.856E+09	0.128E+10	0.171E+10	0.214E+10
1.74	0.974E+09	0.853E+09	0.128E+10	0.171E+10	0.213E+10
		C_0 = 850.0E−03 kg/m^3			
0.99	0.301E+10	0.131E+10	0.197E+10	0.262E+10	0.328E+10
1.24	0.210E+10	0.131E+10	0.196E+10	0.261E+10	0.327E+10
1.49	0.156E+10	0.130E+10	0.195E+10	0.260E+10	0.326E+10
1.74	0.122E+10	0.130E+10	0.195E+10	0.260E+10	0.325E+10

due to the presence of a driving force has been described by taking into account the phenomenon known as concentration-polarization (5, 9, 17) by applying film theory (18).

Every UF process that uses a selective membrane, assumed to be homogeneous and nonporous, includes retainment of solute; thus, an accumulation of solute will unavoidably occur in the neighborhood of and on the membrane surface, resulting in a progressive increase of the solute concentration in this region. This process continues until a certain equilibrium value has been reached. At this point this effect is balanced by diffusion

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of the solute into the bulk of the solution. For most solutions in the UF range, this is considered to be the limiting value of the polarized film or gel concentration, and solute precipitation and membrane fouling can result after this point is reached.

The diffusion model applied here considers Fick's first law (18) by applying a mass balance to the process (6), a balance in the SS in which convective transport and diffusive transport are equilibrated with the mass flux in the permeate as described by

$$J_S = J_V C_A - D_A \frac{dC_A}{d\delta} \tag{6}$$

in which the solute flux in the permeate can be expressed by

$$J_S = J_V C_P \tag{7}$$

so that in integrating Eq. (6) after substituting into it Eq. (7) and taking the limits C_0 for $\delta = 0$ and C_G for $\delta = z$, it follows that

$$J_V = K_M \ln \frac{C_G - C_P}{C_0 - C_P} \tag{8}$$

The observed rejection is defined as a characteristic membrane parameter by the general expression

$$R_{OBS} = \frac{C_0 - C_P}{C_0} \tag{9}$$

For the limit or maximum concentration at the membrane surface, the true rejection can be defined in the same way by

$$R_{MAX} = \frac{C_G - C_P}{C_G} \tag{10}$$

Substitution of Eqs. (9) and (10) into Eq. (8) yields

$$\ln \frac{1 - R_{OBS}}{R_{OBS}} = \ln \frac{1 - R_{MAX}}{R_{MAX}} + \frac{J_V}{K_M} \tag{11}$$

The mass transfer coefficient, which is a function of the velocity, is generally expressed in the form

$$K_M = \frac{D_A}{z} = \phi_D U_0^{\varepsilon} \tag{12}$$

and is used in different generalized correlations (3, 6). The exponent of the velocity takes characteristic values which are functions of the flow conditions and evolutionary steps of the limit layers of velocity and con-

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centration. The constant ϕ_D includes contributions due to the nonuniformity of the diffusion coefficient (to obtain Eq. 8 it has been considered to be independent of the concentration), viscosity, and density as geometric features of the module.

Since knowing the mass transfer coefficient is fundamental in order to predict the behavior of a membrane with respect to the permeate flux, the experimental data were correlated to Eq. (11) by using the procedure described above (14) which uses the method of variation of the circulation velocity and which allows the true solute retention for each concentration considered in this work to be obtained.

For different feed inlet concentrations and velocities of 0.99, 1.24, and 1.49 m/s, corresponding to the transition flow, Grober's equation was used. For a velocity of 1.74 m/s, corresponding to turbulent flow, the Chilton–Colburn correlation was considered.

The results obtained (Table 3) after correlating the experimental data were represented graphically (e.g., Fig. 5), showing that there exists an

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TABLE 3
Results Obtained for the True Rejection and the Mass Transfer Coefficients (Eq. 11)

C_0 (kg·m^{-3})	U_0 (m·s^{-1})	R_{MAX} (% w/w)	C_G(mean) (kg·m^{-3})	e	K_M (m·s^{-1})	F (Eq. 4)
0.425E+01	0.99	92.13	0.765E−01	0.5	0.394E−04	0.98E−01
	1.24			0.5	0.479E−04	0.37E−01
	1.49			0.5	0.694E−04	0.23E−02
	1.74			0.8	0.166E−03	0.54E−02
0.850E−01	0.99	95.52	0.153E+00	0.5	0.362E−04	0.92E−01
	1.24			0.5	0.528E−04	0.17E−01
	1.49			0.5	0.687E−04	0.30E−01
	1.74			0.8	0.119E−03	0.21E−01
0.170E+00	0.99	97.51	0.412E+00	0.5	0.278E−04	0.15E−01
	1.24			0.5	0.310E−04	0.62E−02
	1.49			0.5	0.378E−04	0.25E−02
	1.74			0.8	0.513E−04	0.18E−01
0.425E+00	0.99	98.98	0.145E+01	0.5	0.197E−04	0.25E−01
	1.24			0.5	0.224E−04	0.27E−01
	1.49			0.5	0.226E−04	0.10E−01
	1.74			0.8	0.317E−04	0.34E−02
0.850E+00	0.99	98.98	0.313E+01	0.5	0.169E−04	0.61E−02
	1.24			0.5	0.198E−04	0.63E−02
	1.49			0.5	0.230E−04	0.12E−01
	1.74			0.8	0.266E−04	0.38E−02

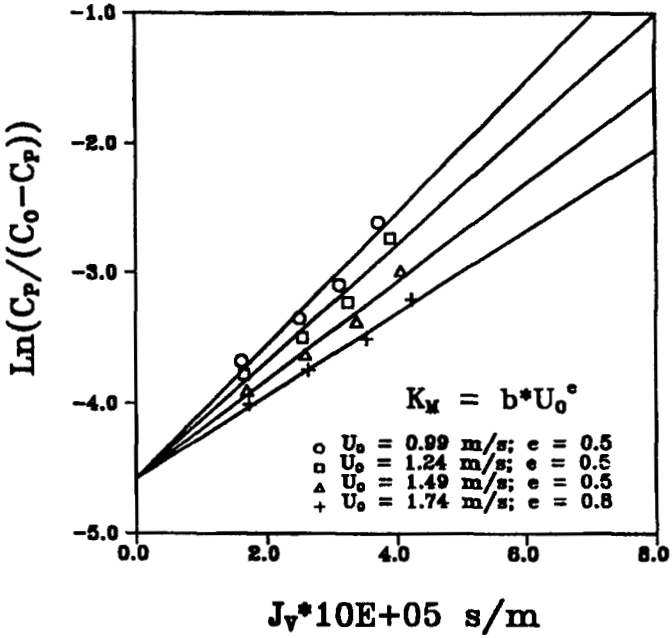

FIG. 5 True rejection for C_0 = 425.0 ppm and evolution of the mass transfer coefficient.

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important influence of the feed concentration on the gel concentration, i.e., on the true retention, and that the mass transfer coefficients show an increasing evolution with respect to the circulation velocity. In order to characterize qualitatively the influence of the velocity on the mass transfer coefficients, the data were plotted (Fig. 6). The differences in dependence of the mass transfer coefficient on the feed inlet velocity at various inlet solute concentrations can be observed in the figure. This dependence is low at high concentrations and high velocities when flow is turbulent.

Several solutions with less than 2% w/w starch/water were prepared in order to obtain their density and viscosity at 298 K. After being correlated, the experimental data gave the following results:

$$\rho = 997.03 + 0.43C_0 - 0.01C_0^2 \quad \text{(standard deviation} = 0.01) \quad (13)$$

$$\mu = (9.10 \times 10^{-4}) + (5.45 \times 10^{-5}\, C_0) + (3.0 \times 10^{-7}\, C_0^2) \quad (14)$$
$$\text{(standard deviation} = 0.13 \times 10^{-4})$$

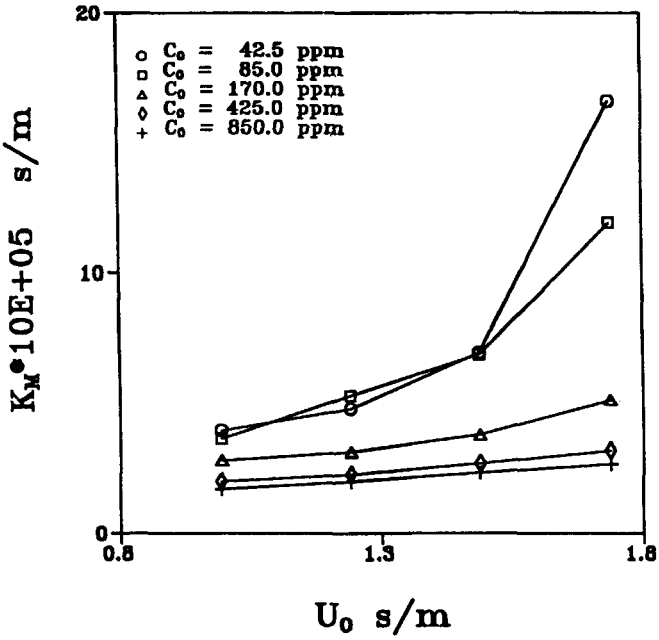

FIG. 6 The influence of the velocity on the mass transfer coefficient.

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The densities and viscosities of the starch solutions considered in this work were determined by means of Eqs. (13) and (14). The results obtained were used in the Grober and Chilton–Colburn equations and gave a diffusion coefficient of $D_A = 1.22 \times 10^{-9}$ $m^2 \cdot s^{-1}$.

CONCLUSIONS

A UF module with plane membranes in crossflow operation was tested in a batch installation with starch solutions. The experimental data obtained after modifying the concentration of the feed solution, the inlet velocity, and the transmembrane pressure have been presented.

After analyzing the behavior of the experimental data, all correlated simultaneously to a series-resistance model, the fouling and gel resistances were obtained. A remarkable relationship between the fouling resistance and the feed inlet velocity was observed, so that increasing the velocity decreases the resistance. Doubling the velocity and a 20-fold increase of feed inlet solute concentration produces no changes in fouling resistance.

Once the contribution of the gel resistance was obtained, assumed compressible, it showed a smaller influence on the velocity than the fouling resistance decrease with increasing velocity. This could be interpreted as a consequence of the stabilization of the gel resistance in the steady state. A linear relationship between the feed inlet solute concentration and the gel resistance was not obtained.

The experimental data were correlated to a diffusion model using the variation of velocity method. This allowed the mass transfer coefficients and the average value of the gel concentration for each of the inlet solute concentrations, which were lower than 5% w/w, to be obtained. A relationship between the mass transfer coefficients and the velocity was observed, so that after determining the densities and viscosities of the different starch solutions, the experimental data corresponding to the transition flow were correlated to the Grober equation, and those corresponding to the turbulent flow were correlated to the Chilton–Colburn equation. In this way the increasing evolution of the mass transfer coefficients with the velocity was proved, and the diffusion coefficient of the solute was obtained.

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ACKNOWLEDGMENT

The authors are grateful to Dirección General XI (Unión Europea) for financial support to LIFE 092-E005 project.

NOMENCLATURE

C_G	limit concentration or gel concentration ($kg \cdot m^{-3}$)
C_0	inlet solute concentration ($kg \cdot m^{-3}$)
C_P	permeate solute concentration ($kg \cdot m^{-3}$)
D_A	diffusion coefficient ($m^2 \cdot s^{-1}$)
J_S	solute flux in the permeate ($kg \cdot m^{-2} \cdot s^{-1}$)
J_V	volume flux ($m \cdot s^{-1}$)
K_M	mass transfer coefficient ($m \cdot s^{-1}$)
L_P	hydraulic permeability coefficient ($Pa^{-1} \cdot m \cdot s^{-1}$)
R_M	membrane resistance ($1/L_P$) ($Pa \cdot m^{-1} \cdot s$)
R_F	fouling layer resistance ($Pa \cdot m^{-1} \cdot s$)
R_G	polarization layer resistance ($Pa \cdot m^{-1} \cdot s$)
U_0	feed inlet velocity ($m \cdot s^{-1}$)
z	thickness of the film layer (m)
ΔP	transmembrane pressure ($\sum P_{inside}/2 - P_{outside}$) (Pa)
ϕ_{RF}	fouling dynamic parameter ($kg^{1-a} \cdot m^{3a-b-2} \cdot s^{b-1}$)
ϕ_{RG}	renovation of mass parameter in gel layer ($kg^{c} \cdot m^{3c-d-1} \cdot s^{1+d}$)
ρ	density of feed solution ($kg \cdot m^{-3}$)
μ	dynamic viscosity ($kg \cdot m^{-1} \cdot s^{-1}$)

REFERENCES

1. V. Gekas, "Terminology for Pressure-Driven Membrane Operations," *Desalination*, *68*, 77–92 (1988).
2. T. D. Brock, *Membrane Filtration: A User's Guide and Reference Manual*, Science Tech. Inc., Madison, Wisconsin, 1983.
3. M. Cheryan, *Ultrafiltration Handbook*, Technomic Publishing Inc., Pennsylvania, 1986.
4. H. M. Yeh and T. W. Cheng, "Resistance-in-Series for Membrane Ultrafiltration in Hollow Fibers of Tube-and-Shell Arrangement," *Sep. Sci. Technol.*, *28*, 1341–1355 (1993).
5. S. Sourirajan, *Reverse Osmosis*, Academic Press, New York, 1970.
6. R. E. Treybal, *Mass Transfer Operations*, McGraw-Hill, New York, 1981.
7. A. G. Bozzano and Ch. E. Glatz, "Separation of Proteins fron Polyelectrolytes by Ultrafiltration," *J. Membr. Sci.*, *55*, 181–198 (1991).
8. G. B. van den Berg, and C. A. Smolders, "Flux Decline in Ultrafiltration Processes," *Desalination*, *77*, 101–133 (1990).
9. G. B. van den Berg and C. A. Smolders, "Concentration Polarization Phenomena during Dead-End UF of Protein Mixtures. The Influence of Solute–Solute Interactions," *J. Membr. Sci.*, *47*, 1–24 (1989).
10. R. M. McDonogh, C. J. D. Fell, and A. G. Fare, "Surface Carge and Permeability in the UF of Non-Floculating Colloids," *Ibid.*, *21*, 285–294 (1984).
11. S. Ilias and R. Govind, "A Study on Concentration Polarization in UF," *Sep. Sci. Technol.*, *28*, 361–381 (1993).
12. R. W. Baker and H. Strathmann, "Ultrafiltration of Macromolecular Solutions with High-Flux Membranes," *J. Appl. Polym. Sci.*, *14*, 1197–1241 (1970).
13. W. F. Blatt, A. Dravid, A. S. Michaels, and L. Nelsen, "Solute Polarization and Cake Formation in Membrane UF: Causes, Consequences and Control Techniques," in *Membrane Science and Technology* (J. E. Flinn, Ed.), New York, 1970.
14. J. Nelder and R. Mead, "A Simplex Method for Function Minimization," *Comput. J.*, *7*, 308–313 (1965).
15. C. E. Reid and E. J. Breton, "Water and Ionic Flow across Cellulosic Membranes," *J. Appl. Polym. Sci.*, *1*, 133–143 (1959).
16. H. K. Lonsdale, U. Merten, and R. L. Riley, "Transport Properties of Cellulose Acetate Osmotic Membranes," *Ibid.*, *9*, 1341–1362 (1965).
17. S. Kimura and S. Sourirajan, "Analysis of Data in Reverse Osmosis with Porous Cellulose Acetate Membranes," *AIChE J.*, *13*, 497–503 (1967).
18. R. B. Bird, W. E. Stewart, and E. N. Lightfoot, *Transport Phenomena*, Wiley, New York, 1960.

Received by editor March 20, 1995
Revision received May 12, 1995

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