INVESTIGATIONS OF HYDRODYNAMIC PERMEABILITY OF CERAMIC AND POLYSULFONE MEMBRANES FOR MICROFILTRATION AND ULTRAFILTRATION

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Abstract: The aim of this research was to compare the hydraulic resistance of several commercial ultrafiltration and microfiltration membranes. The hydraulic resistance for each membrane was calculated from the pure water permeation data collected at various transmembrane pressures and temperatures to check the effects of these parameters on the membrane resistance. The experiments have been carried out in a laboratory crossflow UF/MF equipment for clarification of fruit juices. This paper introduces the experimental results showing the influence of operating parameters, such as feed flow rate, temperature, pressure difference in the microfiltration and ultrafiltration through the ceramic Kerasep membrane (pore size 0.2 μm, Tech-Sep, Miribel, France), Carbosep M9 and M7 membrane (molecular weight cut-off of 300 and 30 kg/mol, Tech-Sep, Miribel, France) and polysulfone membranes (molecular weight cut-off of 30 kg/mol, Frenesius, Germany). The results confirmed our earlier work [1] but we provide here many additional results.

The hydraulic resistance R_m was 0.65, 3.56, and 0.05 × 10^{13} 1/m for Carbosep M7, Carbosep M9 and Kerasep membrane, respectively. The hydraulic resistance of these membranes does not depend on the operating pressure, which means that the membranes are incompressible.

The hydraulic resistance of a polysulfone hollow fiber membrane slightly increased with increasing the applied pressure difference. The R_m values were in the range of $(0.61 - 0.92) \cdot 10^{13}$ 1/m, and the applied pressure difference was in the range of (0.225 – 0.900) bar, which is an indication that this membrane was compressible.
Key words: microfiltration, ultrafiltration, hydraulic resistance, membrane.

INTRODUCTION

Microfiltration (MF) / ultrafiltration (UF) is a membrane separation process which enables the separation of macrocolutes in the range of 0.1 to 10 μm (for MF) / 1 to 100 nm (for UF) from the solvent and other smaller constituents with driving force ranging from 1 to 5 bar. The concept of membrane separation processes is in its essence very simple. Using an appropriate driving force, all components that cannot pass through the membrane are retained in the feed solution, whereas the other components are removed by permeation throughthe membrane . In MF and UF the solvent molecules are transported through the membrane pores by convective flow, and suspended particles are retained on the membrane surface just like on a sieve, because the membrane pores are too narrow for the particles to pass through.

Compared to the conventional processes, MF / UF can bring the following benefits to the processors: separation can be carried out without changing temperature and pH of solution and without adding chemicals such as fining agents, the production costs and the problem of waste water treatment are reduced, the product quality is improved and the labor costs are lower.

MF and UF can find applications in the food and beverage industry for fruit juices clarification and recovery of valuable products from the waste streams such as colorants.

The volumetric rate of flow, Q_V (m^3/s), for the permeation of pure water of viscosity, μ (Pa·s), through an isotropic membrane of thickness δ (m), pore diameter d_p (m), and tortuosity factor ξ under the applied pressure difference Δp (bar) is given by:

$$Q_v = vA_o = \frac{\Delta p d_p^2 A_o}{32 \mu \delta \xi} = JA_m = \frac{\Delta p A_m}{\mu R_m} \qquad (1)$$

where v is the average velocity of permeate through the pores, A_o is the mean cross-sectional area for flow, A_m is the effective cross-sectional membrane area, J is the permeate flux, R_m is the hydraulic membrane resistance.

Therefore, the permeate flux of pure water through the membrane can be written as:

$$J = \frac{Q_v}{A_m} = \frac{Q_m}{\rho A_m} = \frac{\Delta p}{\mu R_m} \qquad (2)$$

where Q_m is the mass flow rate of permeate and ρ is permeate density. It follows from Eq. (1) that the hydraulic resistance of isotropic (symmetric) membrane can be represented by:

$$R_m = \frac{\Delta p A_m \rho}{\mu Q_m} = \frac{32 \delta \xi}{d_p^2 \varepsilon} \qquad (3)$$

where $\varepsilon = A_o/A_m$ is the membrane porosity. Because the hydraulic resistance of the membrane is directly proportional to the membrane thickness and inversely proportional to the square of the pore diameter, all comercial UF and MF membranes are anisotropic in structure. The anisotropic membrane posses a thin permselective layer of thickness δ' and pore diameter d_p' on a porous support layer of thickness δ'' and pore diameter d_p''. Since the resistances of skin and support layer are connected in series, the total resistance of anisotropic membrane is as follows:

$$R_m = R_m' + R_m'' = \frac{32 \delta' \xi'}{d_p'^2 \varepsilon'} + \frac{32 \delta'' \xi''}{d_p''^2 \varepsilon''} \qquad (4)$$

As an example, if $\delta' = 0{,}5$ μm, $\delta'' = 220$ μm, $d_p' = 4$ nm, $d_p'' = 1$ μm and $\xi'/\varepsilon' = \xi''/\varepsilon''$, then from Eq. (4) $R_m' = 142 R_m''$, i.e. in spite of 440 times greater thickness the resistance of support layer can be neglected compared to the resistance of dense permselective layer. The thickness of an isotropic membrane is inevitably greater than thickness of selective layer of an anisotropic membrane and therefore, at the same separation abilities and operating conditions anisotropic membranes enable much higher pure solvent fluxes than isotropic membranes.

MATERIAL AND METHODS

The experiments were caried out on the laboratory device for microfiltration and ultrafiltration. Laboratory device for MF and UF is designed to enable easy handling, process verification and changing of operating parameters (flow of distilled water through the module, pressure difference,

temperature). The shematic view of the experimental equipment used for the measurement of distilled water permeabilites of MF / UF membranes is shown in fig.1.

The distilled water was pumped from the reservoir to the module by a rotary pump and flow rate was monitored with a laboratory made rotameter. Temperature in the system was adjusted by passing water from a bypass line through the thermostat bath. Permeat was collected in a graduate cylinder placed on a Tehtnica model ET-111 digital balance. Overpressure in the system was adjusted by back-pressure regulator and measurement was done with a pressure gauge.

The experiments were performed with inorganic microfiltration ceramic Kerasep membrane (pore size 0.2 μm, Tech-Sep, Miribel, France), inorganic ultrafiltration membranes Carbosep M9 and M7 (molecular weight cut-off of 300 and 30 kg/mol, Tech-Sep, Miribel, France) and organic ultrafilration membrane of a polysulfone hollow fiber (molecular weight cut-off of 30 kg/mol, Frenesius, Germany). Ceramic Kerasep membrane has 19 channels with 4 mm diameter, effective lenght 270 cm and effective membrane area of 0.0644 m^2. This membrane was installed inside a plastic module with a stopper tire. Ultrafiltration membranes Carbosep M9 and M7 are composed of thin permselective skin of zirconium oxide and titanium dioxide supported by a porous carbon substructure. The membranes were installed inside a cylindrical stainless steel module with an effective membrane lenght of 225 mm and an effective membrane area of 42.4 m^2. Polysulfone hollow fiber membrane is made from polysulfone with designation ultraflux AV1000S. This membrane has 10258 fibers with internal diameter of 220 μm and wall thickness of 35 μm. They were installed inside a polycarbonate module with a polyuretane stopper material. Effective lenght of this membrane is 254 mm and an effective membrane area is 1.8 m^2.

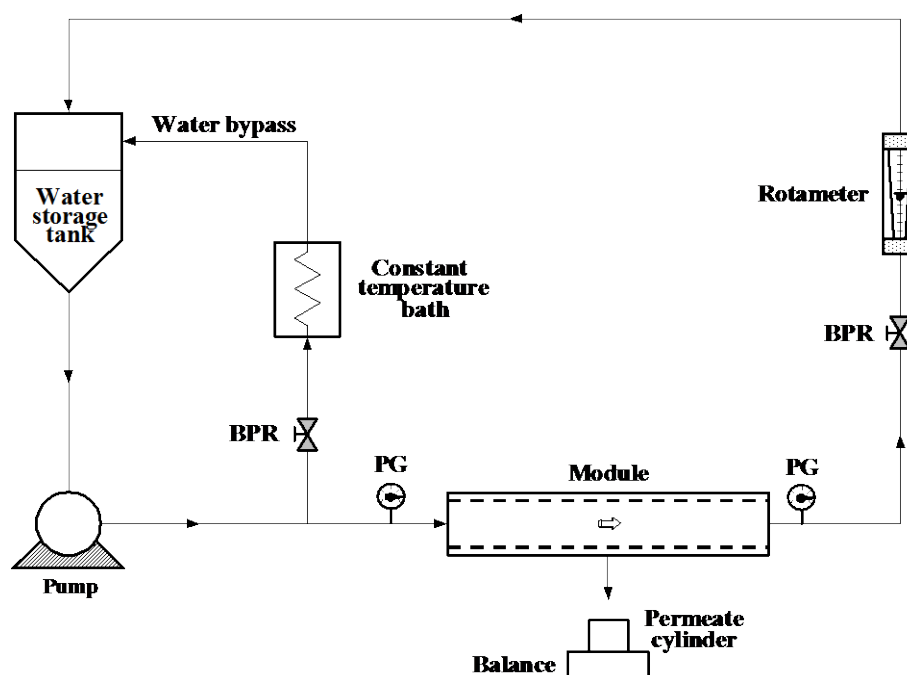The results confirmed our earlier work [1] but we provide here many additional results.

Fig. 1. Scematic view of the experimental setup employed for the measurement of pure water permeabilities
(PG-pressure gauge, BPR-back-pressure regulator)

RESULTS AND DISSCUSION

The hydraulic membrane resistance, R_m, has been determined by the distilled water permeation through the membranes at the applied pressure difference in the range from 0.5 to 3 bar (for Carbosep M7 and M9 membranes), 0.5 to 1.5 bar (for Kerasep membrane) and 0.225 to 0.900 bar (for polysulfone membranes), temperature of 22, 35, 45 and 55°C and feed flow of distilled water throught the module Q_v = 1 l/min. The hydraulic resistances of the membrane can be calculated from Eq.(3) using the permeate flow rate values determined by the least-squares regression analysis method. Time of one experiments was 120 min. In fig.2 and 3 is shown the effect of applied pressure difference and temperature on the hydraulic membrane resistance, with feed flow rate of 1 l/min, for above mention membranes in the text, and the hydraulic resistances, R_m (1/m) for several commercial UF and MF membranes.
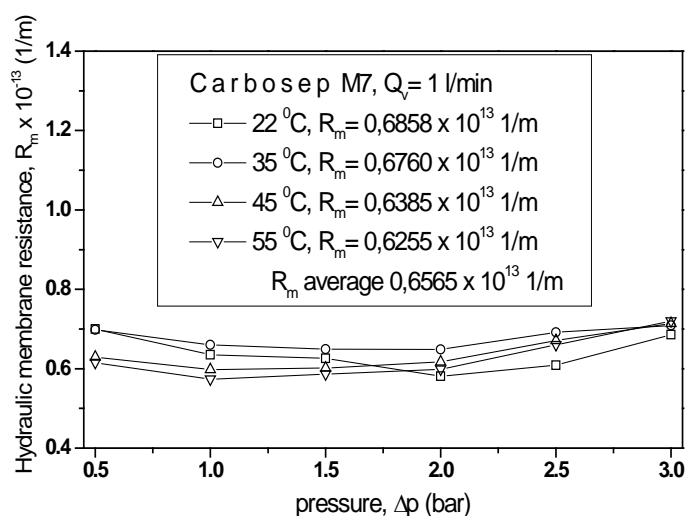

Fig.2. Influence of pressure difference, Δp (bar), and temperature, t (°C), on the hydraulic membrane resistance, R_m (1/m), for Carbosep M7 membrane with retentate flow Q_v = 1 l/min

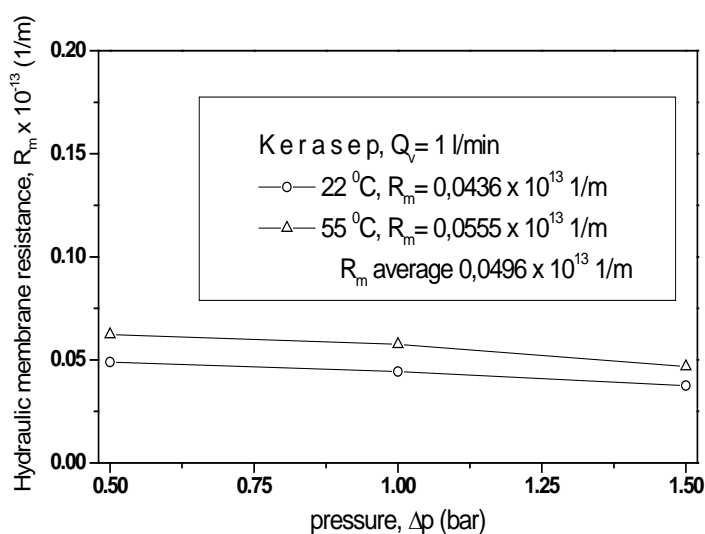
Fig.3. Influence of pressure difference, Δp (bar), and temperature, t (°C), on the hydraulic membrane resistance, R_m (1/m), for Kerasep membrane with retentate flow Q_v = 1 l/min

Fig.4. Influence of pressure difference, Δp (bar), and temperature, t (°C), on the hydraulic membrane resistance, R_m (1/m), for polysulfone membrane with retentate flow $Q_v = 1$ l/min

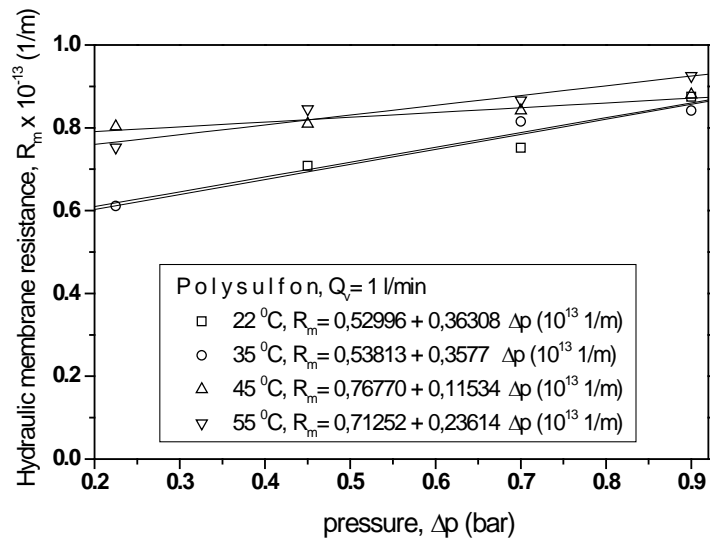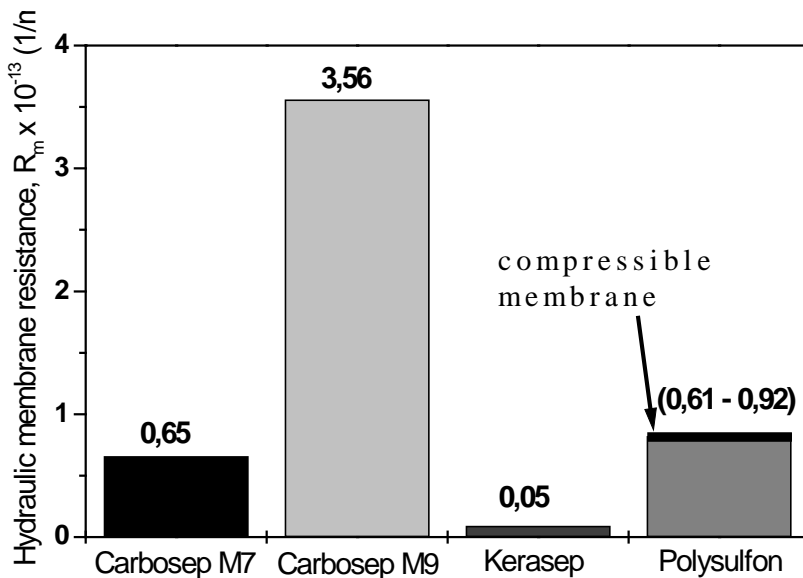
Fig.5. The hydraulic resistances, R_m (1/m) for several commercial UF and MF membranes

The hydraulic membrane resistance (R_m), for the Kerasep membrane, Carbosep M7 and Carbosep M9 membranes was independent on the applied pressure difference, temperature and feed flow rate. The R_m is constant value, which means that the membranes are incompressible. The R_m value were $0.65 \cdot 10^{13}$ 1/m for Carbosep M7 membrane, $3.56 \cdot 10^{13}$ 1/m for Carbosep M9 membrane and $0.05 \cdot 10^{13}$ 1/m for Kerasep membrane (fig.3). The hydraulic resistance of a polysulfone hollow fiber membrane slightly increased with increasing of the applied pressure difference (fig.4). The R_m

value were in the range of $(0.61 - 0.92) \cdot 10^{13}$ 1/m which means that this membrane was compressible (fig.5).

The transmembrane flux of distilled water depended on the transmembrane pressure, temperature i.e. water viscosity and the hidraulic resistance.

CONCLUSIONS

The results of the performed experiments, where investigation was carried out on the influence of operating conditions (pressure difference, temperature, flow of distilled water through the module) on MF and UF of distilled water through the several commercial UF and MF membranes were used to determined hydraulic resistance of that membranes. The hydraulic resistance of Carbosep M7 and M9 and Kerasep membrane is independent on operating conditions. R_m is constant value and dependent onla on membrane type. The hydraulic resistance of polysulfone membrane is dependent on pressure difference (driving force) and slightly increased with pressure, because this membranes are compressible.

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