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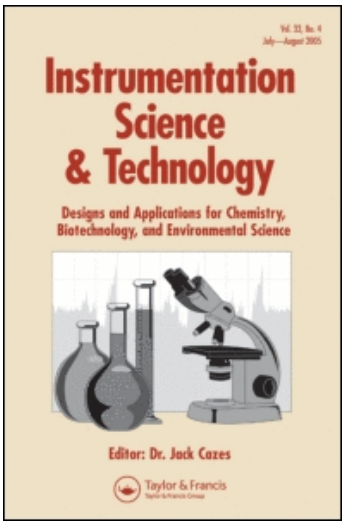

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An Apparatus for Automated Cross Flow Solute Permeation Characterization of Membranes

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Abstract: An apparatus is described for the automated characterization of ultrafiltration membranes using solute permeation in cross flow mode. The automated characterization approach described in this work lends itself well for the purpose of increased productivity and reducing operator fatigue/error. The operational, control, and data acquisition aspects of an automated membrane cross flow test unit, which are accomplished using LabVIEW 5.0™ are described. The interpretation of the flux and separation data is independent of the apparatus and depends on the filtration regime and various theoretical models available. The apparatus can be used for reverse osmosis, nanofiltration, or ultrafiltration experiments, with appropriate selection of test cells and pumps.

Keywords: Membrane, Characterization, Permeation, Sieving, Cross flow, Separation

INTRODUCTION

Discovery of asymmetric membranes by Loeb and Sourirajan for reverse osmosis in 1959 initiated the modern era of membrane based separations.[1]

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The asymmetric nature of phase inversion membranes increased permeation fluxes to the level of commercial viability. Improvements in membrane performance are ongoing with the development of new membrane materials, preparation techniques, casting formulations, and post processing methods. Reliable, accurate, and quick methods to characterize membrane performance, as measured by flux and separation characteristics, play a crucial role in this development. Several complementary methods such as: microscopic techniques (scanning electron microscopy (SEM), atomic force microscopy (AFM)), used alone or in conjunction with computer aided analysis,[2] solute permeation experiments, liquid-liquid displacement porosimetry (LLDP), and bubble point, are available for membrane characterization. All of these techniques have some inherent limitations and membranes are often characterized by more than one method.

The SEM resolution of uncoated polymeric membranes is limited by the beam energy, such that the polymer would not be degraded, or to prevent charge accumulation. Inorganic membranes can be imaged well using SEM since they do not have charge accumulation or degradation issues. An AFM with a liquid cell is well suited to polymeric membranes as they do not require drying or gold coating, which may alter the morphology of the membrane.

Flow through porosimetry methods use a non-wetting phase to displace the fluid from prefilled pores. These methods have the advantage of providing a direct pore size distribution. Pore radii of greater than one micron are generally characterized by bubble point techniques using air and water with an interfacial tension of $72\,\mathrm{mPa \cdot m}$. Smaller pore radii would require excessively high pressures, which could result in membrane compaction. LLDP uses co-saturated immiscible phases (typically water, methanol, and iso-butanol mixtures) with lower interfacial tensions ranging from 1.85 to $0.35\,\mathrm{mPa \cdot m}$, allowing measurement of smaller pores at acceptable pressures. A drawback is the potential interaction of the test fluids with the membrane materials resulting in swelling or contraction. The lower limit of LLDP, which is not clearly defined, is for pore radii less than 0.5 nm, below which it is suspected, but not proven, that separation of the fluid mixtures may occur. Some experimental considerations for this technique are discussed.[3]

Solute permeation is a logical methodology to characterize membranes, since the primary task of membranes is to affect a separation. Major criticisms of this approach have been the dependence on the test cell/membrane configuration and interactions between solutes and membrane materials. The test cell dependence can largely be accounted for using the film theory for ultrafiltration membranes via the mass transfer co-efficient and the flux. Solute membrane interactions should be avoided at all costs as they may change the apparent pore size via adsorption, pore blockage, or coating the membrane, depending on the relative pore and solute sizes.[4] Sieving experiments are also time consuming and repetitive, requiring several permeation experiments using different sized solutes to obtain a full sieving curve, often referred to as a Molecular Weight Cut-Off (MWCO) curve.

An apparatus, henceforth called the Automated Permeation Unit or APU, was fabricated, assembled, and programmed in our laboratory to increase the productivity of membrane characterization. The APU characterizes membranes with 1) an initial pure water flux after a pre-set compaction period, 2) on-line recording of trans-membrane pressure, fluid temperature, and permeation rates, and 3) feed and permeate sample collection for each solute for further analysis. All operations such as test-cell and permeate line flushing, solute selection, feed tank washing are automated. Variations in membrane fluxes and separation characteristics are well recognized, if not documented in the literature. The APU characterizes 12 membrane samples simultaneously; this is a significant advantage when developing new membranes, carrying out quality control in production, or pre-screening membranes for application development. The APU greatly reduces operator fatigue and error, and increases throughput. The need for such automation was previously recognized in our laboratories,[5] and recently, the level of automation and data recording has been improved. In particular, variations in temperature and pressure were not recorded to correct data to standard conditions. On-line monitoring of the permeation rate also allows verification of steady state with respect to the membrane flux, and does not require operator input to set sampling or flush times. Other forms of membrane characterization are also being automated, such as screening with stirred cells and a single solute[6] and diffusion through films.[7]

EXPERIMENTAL

Feed Materials and Analysis

Water soluble polyethylene glycols (PEG) from Fluke are available in molecular weights of 200, 600, 1,200, 2,000, 3,000, 6,000, 10,000, 12,000, 20,000, and 35,000 Da, amongst others, with low polydispersity, typically with $Mw/Mn < 1.1$. The low polydispersity allows evaluating separations by total carbon (Shimadzu TOC 5000 with ASI auto sampler). Polyethylene oxides are available in molecular weights of 100 K, 300 K, 600 K, and 900 K. Nanofiltration membranes are characterized using various salts in addition to PEGs. Reverse Osmosis permeate (conductivity less than 10 μS) is used at all stages of characterization and rinsing. Solutes are generally permeated in order of increasing molecular weight and as solutions of one molecular weight, mixtures were shown to bias the separations.[8]

Permeation Cells

Cross flow permeation cells of any geometry and size can be used, with suitable consideration to pump sizes. Figure 1 illustrates the cross sectional

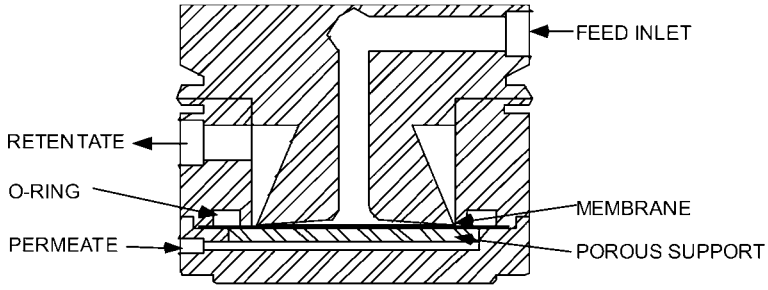

Figure 1. Schematic of permeation cell.

view of a typical permeation cell used in our laboratory. In this cell, the retentate flows radially outwards from the axial center. The gap at the circumference is 5.1×10^{-4} m with a permeation area of $1.4 \times 10^{-4}\,m^2$. The typical volumetric flow rate for the cell is $3\,L \cdot h^{-1}$, yielding a cross flow velocity of $0.8\,m/s$ at the outer edge of the cell. An unusually high flux of $1{,}000\,L \cdot m^{-2} \cdot h^{-1}$ represents $\sim 0.8\%$ of the retentate; an insignificant amount.

Data Analysis

The observed separation, f_{obs}, is determined from, Equation (1) where C_{feed} and C_{perm} are the feed and permeate concentrations, respectively.

$$f_{obs} = \frac{C_{feed} - C_{perm}}{C_{feed}} \tag{1}$$

During the filtration process, the solute is transported to the membrane surface by convection (i.e., the permeate) and away from the surface by diffusion, D, of the solute across a concentration boundary layer of thickness δ. There is an accumulation of solute at the membrane surface creating a wall concentration, which is greater than the feed concentration. The intrinsic separation of the membrane is given by Equation (2) where C_{wall} is the concentration at the membrane wall:

$$f_{in} = \frac{C_{wall} - C_{perm}}{C_{feed}} \tag{2}$$

The effects of concentration polarization can be accounted for based on the flux and mass transfer using the film theory,[9] Equation (3), where the mass transfer co-efficient, k, is D/δ and J_v is the permeate flux:

$$f_{in} = \frac{1}{1 + ((1 - f_{obs})/f_{obs}) \times \exp(-J_v/k)} \tag{3}$$

The ratio of flux to mass transfer coefficient (J_v/k) can be estimated during the pressurization stage. The operator then adjusts the pressure in that bank (see next section) to reduce the permeation rate to give acceptable values of J_v/k. Lower values of J_v/k are preferred so that the observed separation is closer to the intrinsic separation. However, values <1 may be difficult to achieve for higher molecular weight solutes, in these cases one can only minimize J_v/k.

There are numerous interpretive models in the literature, as starting points: for ultrafiltration,[10] nanofiltration,[11] and reverse osmosis,[12] the later reference discussing the two major approaches to the mechanism of reverse osmosis.

AUTOMATED PERMEATION UNIT

Hydraulic Circuit

The hydraulic circuit for the APU is shown in Figure 2. A total of 12 permeation cells are used, arranged in 4 banks of 3 cells in series. This configuration was selected to minimize the pressure drop across a given bank ($\sim$13 kPa with water at room temperature at 0.8 m/s cross flow velocity). Each bank of permeation cells has: a back pressure valve after the 3rd cell, rotameter, and a magnetically coupled vane pump, with bypass, to allow setting the cross flow velocity and pressure independently. Cartridge prefilters (0.2 μm) are located at the pump outlet to trap trace particulates, followed by a coil in a temperature controlled bath.

All pumps are fed from a common 20 L feed tank. The feed can be pure water (reverse osmosis permeate) or a solution of 1 of 5 possible solutes. The solutions are prepared by diverting the pure water supply through one of the 5 solute bottles during a tank filling sequence. Fluid supply to the feed tank is via a flat, circular spray nozzle to wash the tank walls without completely filling it.

Permeate from each test cell goes to an individual 4-way solenoid valve (Nautilus Research, HBPT-062) sending permeate to: the drain (during flushing stages), the on-line flow meter, or to the sample tray.

Sample Collection

Permeate samples are collected in a tray with a Cartesian arrangement: 8 rows with 14 vials, 1 vial per permeation cell and two vials for the feed sample, one at the beginning and one at the end of the permeation test. Collection times are determined by the on-line flow meter to deliver 15 mL to each vial. Each row is for: an initial pure water flux, solutes 1–2, an intermediate pure water flux, solutes 3–4, a final pure water flux, and solute 5. Permeation times for sample collection are recorded for calculating fluxes as a back up to the on-line flow meter. The low pressure side of the permeation cell and all lines are flushed before sample collection.

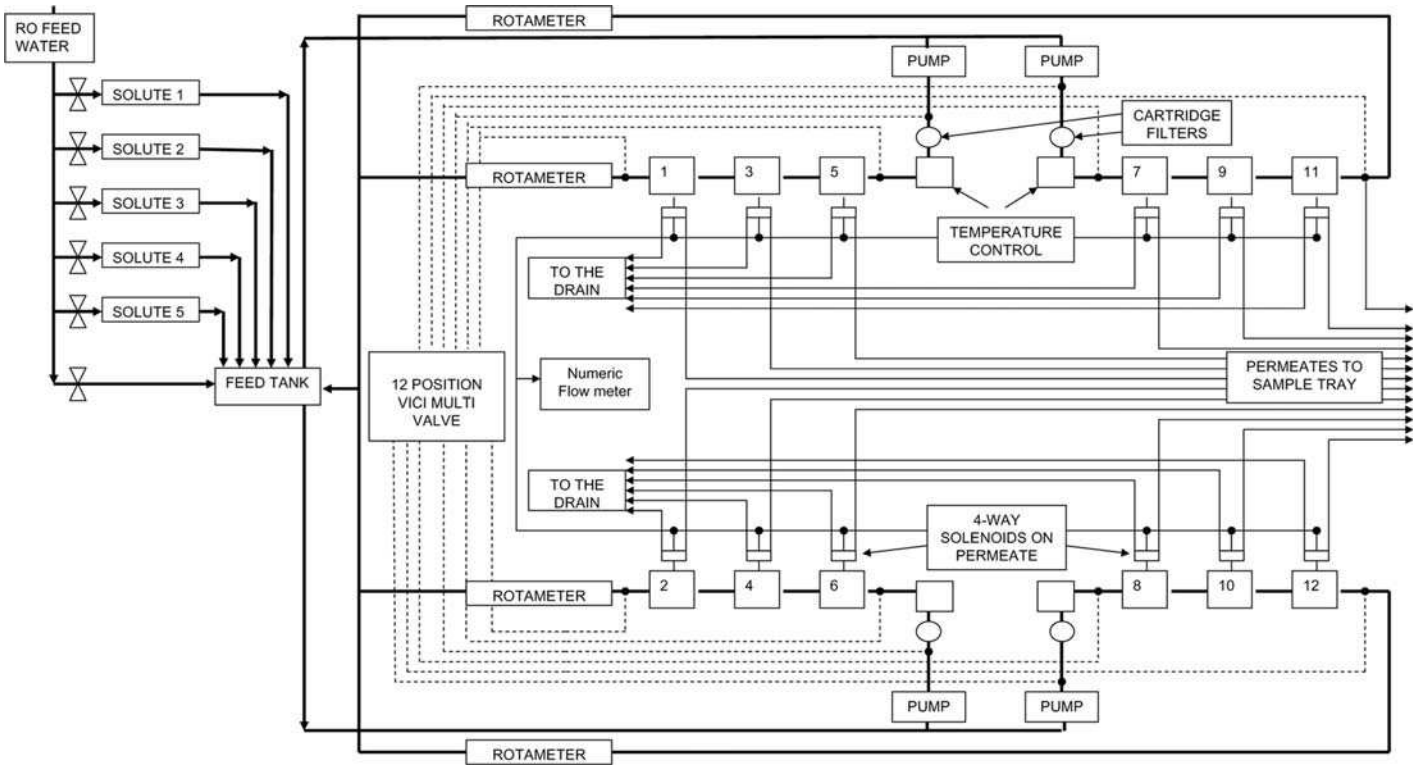

Figure 2. Hydraulic circuit; heavy solid lines are recirculation, light solids are permeate, dashed lines are pressure taps.

Permeate and feed sample lines to the sample tray are located on a support bar using Swagelock™ fittings in a linear array. The "home" position is over a trough, which goes to the drain during permeate sample line flushing. Stepper motors position the sample line array vertically and horizontally above the sample tray. The location of each row is determined in a LabVIEW™ program, which guides the operator through the procedure in a one time setup. In the event of an abnormal shut-down or interrupt, the permeate sample array is returned to the home position in "safe mode"; the sample line array is brought to the upper position and returned to the home position in 1/4 turn steps, using the limit switches to locate the travel limits. The wiring diagram for the stepper motors (Warner Electric, SLO SYN M061-FD-311), interface (Ontrak Control Systems ADR 2100), and driver (Warner Electric, SS 200-DP4) is shown in Figure 3.

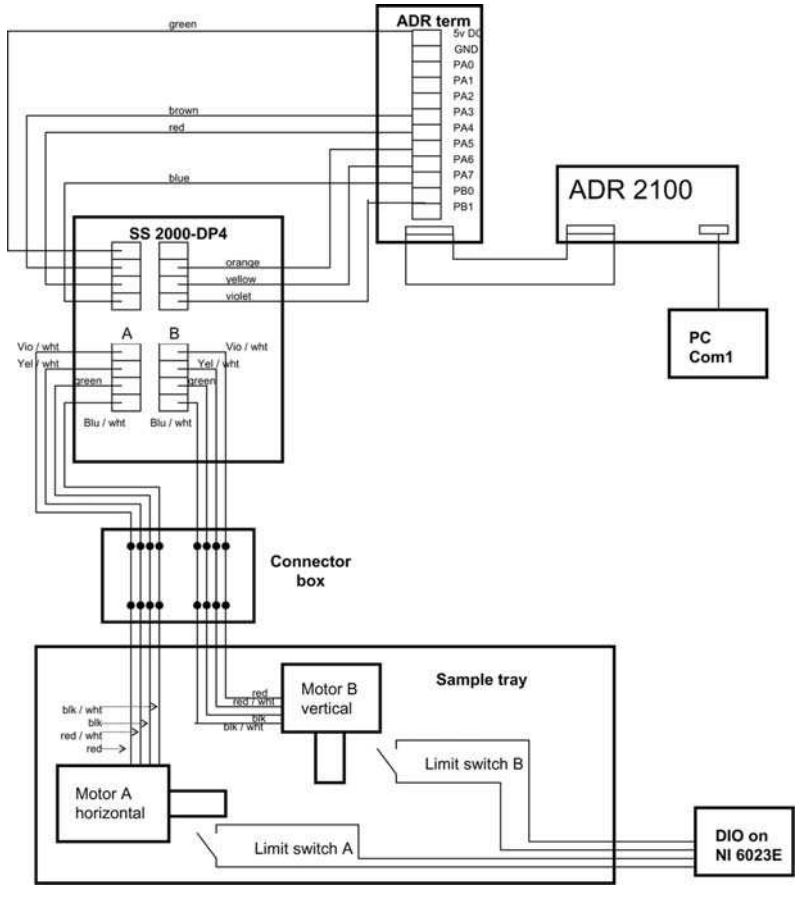

Figure 3. Wiring diagram for stepper motors on sampling collection tray.

Flux Measurements

Volumetric permeate flow rates are measured using a GJC Instruments flow meter, which times the passage of the fluid meniscus between two sensors. Two models are available GJC10001 and GJC10003, with ranges of 0.1–65.5 mL/min and 0.02–10 mL/min, respectively. The former is primarily used in the APU with the permeation cells described above, while the latter is better suited for lower permeation rates, which could result from lower trans-membrane pressures, smaller permeation areas, or low permeability membranes. Flow meter data is acquired via a parallel port. Occasional "data splitting" has been observed when reading flow rates; a number such as 12.345 can appear as 1 and 2.345, 12, and 0.345, 12, and 345, and so on. This was corrected for most occurrences by reading the port two times in rapid succession (less than 0.1 seconds), if two values are present then the data was split, and discarded. If only one value is present, it is correct.

The recorded flux is the average value of the last n measurements of the permeation rate, PR, for which the 95% confidence interval is less than a given fraction, tol, of the average value. The 95% confidence interval is 2 times the product of the current standard deviation, σ, the current average and the $t_{n\text{-}1,0.05/2}$ statistic for the $n\text{-}1$ degrees of freedom associate with σ.

$$\frac{2 \times \sigma \, t_{n-1,0.05/2}/\sqrt{n}}{(1/n \sum_{x}^{x+n} PR)} \leq tol \tag{4}$$

When this condition is met, the next cell is selected for measuring the flux. A typical value for tol is 5% and n is usually set to 6. The F statistic for smaller values of n becomes unduly large, and there is a chance that the criteria will not be met due to random fluctuations in the flow readings.

Test cells are never dead ended; the circulation time serves two purposes, to flush permeate lines of water or the previous solute, and to attain steady state with respect to the formation of the concentration boundary layer at the membrane surface. Although it was shown that fluxes can achieve a steady state in 20–200 seconds,[10] this could represent a significant portion of the sample collection time and, therefore, bias the backup permeation rate measurement. Furthermore, stopping the permeate flow would stop the convective transport of the solute to the membrane surface. The period with no trans-membrane pressure and the flow instability during the initial pump restart up would reduce the solute concentration at the membrane surface. Fluxes and separations would be marginally higher than at steady state.

Temperatures are recorded during the measurement of each cell's permeation rate. Pressures at each bank inlet and outlet are recorded at the beginning of the flow measurement for each bank of cells. Fluxes are normalized for the permeate viscosity and corrected to a standard trans-membrane pressure. Note that the actual flux should be used in Equation (3).

Data Acquisition and Control

Data acquisition is via a National Instruments PCI-6023E (Device 3) with 12 bit resolution for the temperature (Omega DP116-KC2 with K thermocouple) and pressure (transducer from Honeywell SA 0-200 PSI and Precision Digital readout). The card also serves as the digital input/out (DIO), Figure 4, for the sample tray limit switches, a feed tank level switch, and a trigger for the 12 position sample valve for pressure readings. The stepper motors are controlled via a serial port.

The remaining control functions are via two KPCI-PIO-24 relay boards (Device 1 & 2) switching mechanical relays on two ERB-24 electromechanical relays boards (Keithley Instruments). The wiring diagram is shown in Figure 4, with DIO assignments for all three devices summarized in Table 1.

Typical Control Sequence for a MWCO

The primary task of the APU is to generate a MWCO curve and record fluxes. A typical characterization begins with a pressurization stage where pure water

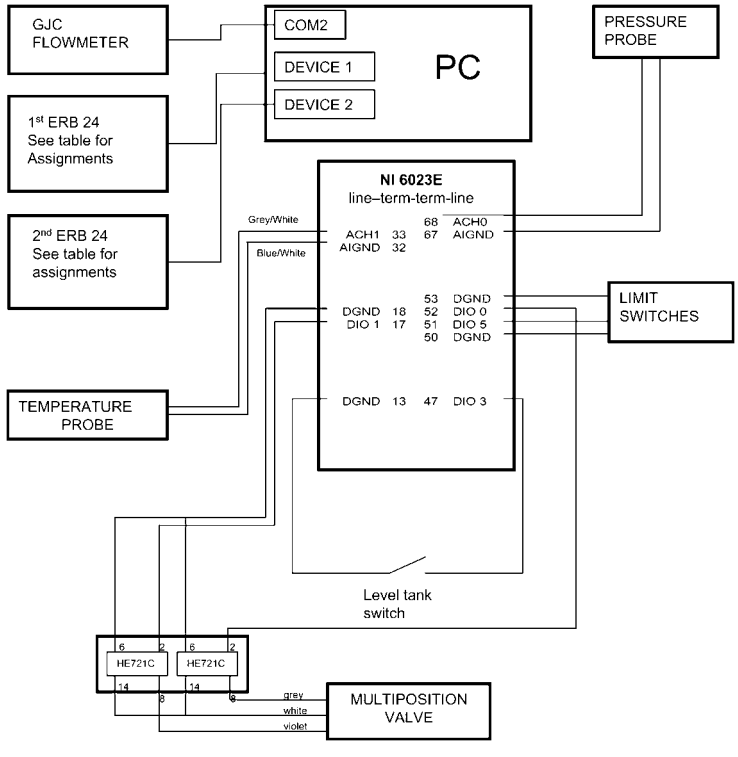

Figure 4. Controls and data acquisition for the automated permeation unit.

Table 1. Digital I/O assignments for ERB-24 boards and 6023E DAQ card

C DIO 24 reference	Address in decimal	Relay number	Upper board = Device 1	Lower board = Device 2
Port A or 0,	1	0	Cell 1, perm to sample	Cell 2, perm to sample
	2	1	Cell 3, perm to sample	Cell 4, perm to sample
	4	2	Cell 5, perm to sample	Cell 6, perm to sample
	8	3	Cell 7, perm to sample	Cell 8, perm to sample
	16	4	Cell 9, perm to sample	Cell 10, perm to sample
	32	5	Cell 11, perm to sample	Cell 12, perm to sample
	64	6		Feed initial
Port A or 0,	128	7		Feed final
Port C or 2,	1	8	Cell 1, perm to drain	Cell 2, perm to drain
	2	9	Cell 3, perm to drain	Cell 4, perm to drain
	4	10	Cell 5, perm to drain	Cell 6, perm to drain
	8	11	Cell 7, perm to drain	Cell 8, perm to drain
	16	12	Cell 9, perm to drain	Cell 10, perm to drain
	32	13	Cell 11, perm to drain	Cell 12, perm to drain
	64	14	Feed, bottle 1	Feed, bottle 4
Port C or 2,	128	15	Feed, bottle 2	Feed, bottle 5

Port B or 1,	1	16	Cell 1 to flowmeter	Cell 2 to flowmeter
	2	17	Cell 3 to flowmeter	Cell 4 to flowmeter
	4	18	Cell 5 to flowmeter	Cell 6 to flowmeter
	8	19	Cell 7 to flowmeter	Cell 8 to flowmeter
	16	20	Cell 9 to flowmeter	Cell 10 to flowmeter
	32	21	Cell 11 to flowmeter	Cell 12 to flowmeter
	64	22	Feed, bottle 3	Distilled water
Port B or 1,	128	23	Pumps	Drain main tank

NI 6023E	Instruments	Type	Channel	Connector numbers
	Step motors limit switches	Digital	0 & 2	49, 50, 52
	Level tank switch	Digital	3	13, 47
	Multiposition valve	Digital	1 & 5	17, 18, 51
	Pressure probe	Analogue	0	67, 68
	Temperature probe	Analogue	2	33, 66
Serial port 1	Stepper motors			
Serial port 2	Flow meter			

is permeated for an extended period (usually 5 h) to compact the membrane and/or remove any solvents/additives used during membrane preparation or preservatives used for long term membrane storage. The sequence for the permeation of solutes and intermediate pure water fluxes is as previously described for the sample tray. The procedures for pure water and solute permeations differ only during the tank filling step, when fresh RO water is directed through one of the five solute reservoirs (Figure 2).

A brief summary of the control sequence for a pure water or solute permeation test, shown in Figure 5, was implemented by developing computer programs under the LabVIEW^{TM} programming environment. LabVIEW^{TM} software is a graphical development environment for scalable test, measurement and control applications. Each block represents a sequence of events controlled by subroutines; Virtual Instruments (VIs) in

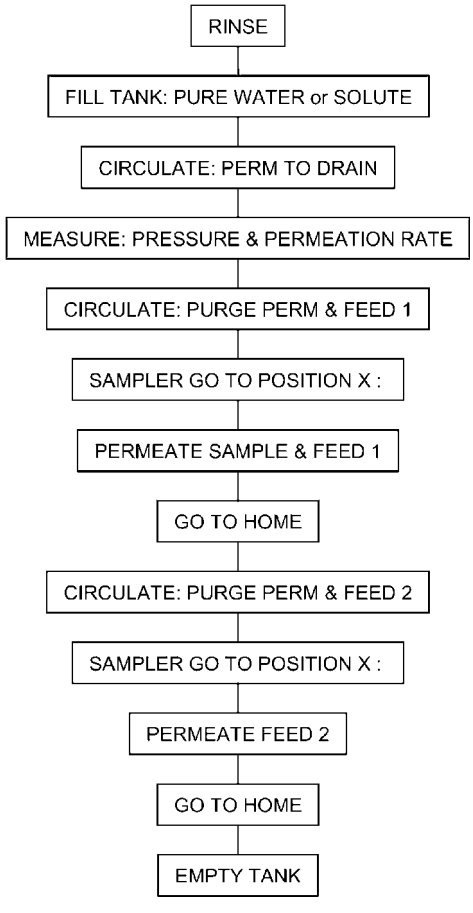

Figure 5. Action sequence for preparing and collecting a solute.

LabVIEW^{TM} nomenclature. For example, the RINSE routine rinses the feed tank with fresh RO water while the drain is open. The drain is then closed, the tank partially filled, the pump turned on and permeate directed, alternately, to the drain or the sampling needles, for 5 cycles. The RINSE subroutine is intended to flush the system of the previous solute.

The CIRCULATE subroutine is similar to RINSE but its function is to ensure that the permeate side of the cells and the lines going to the sample tray are filled with permeate containing the steady-state concentration of solute. Manual tests have shown that permeating 15 mL of liquid is sufficient to achieve a steady-state permeate concentration.

Time savings are significant: a typical MWCO experiment on the APU can be done in 24–36 hrs versus an intensive 3–4 day test on a manual system. Actual tests may take longer if permeation rates are extremely low and flush times take longer. In these cases, the benefits of the APU are increased as sample collection is not restricted to normal working day hours. It is not our intent to display all the block diagrams in this article; a copy of the full set of VI's and equipment list is available to anyone by contacting the corresponding author.

There are several pre-programmed sequences allowing the operator to: 1) perform a MWCO, 2) calibrate sampler tray positions, 3) empty and rinse the feed tank, 4) fill the feed tank, 5) circulate the feed, and 6) pressurize/compact the membranes.

Some of these stand alone functions are part of a normal MWCO, but are often used alone during maintenance procedures. Calibrating the sampler tray positions is rarely repeated.

RESULTS

MWCO Examples

In an example illustrating the use of the APU as a developmental tool; twelve samples were cut from a 1 m × 2.5 m polyethersulfone membrane sheet prepared in our laboratories. Samples were taken at each edge and the center; at 0.25, 1.38, 1.75, and 2.16 m. The intrinsic MWCO curve for the samples is shown in Figure 6. The solute molecular weight with 90% rejection is typically used to describe the MWCO of a membrane; the range of MWCO's was approximately 2 to 4.5 kDa for this particular casting. The variation of the initial pure water flux, corrected to 340 kPa trans-membrane pressure and 25°C, is minor, averaging $62.3 \pm 3.5\,\mathrm{L \cdot m^{-2} \cdot h^{-1}}$ at the 95% confidence level. Based on previous experience with commercial membranes, these ranges for the pure water flux and MWCO are very acceptable.

Quality control is important for cases where membranes are used to fractionate or recover high value added products. Ten samples of a commercially

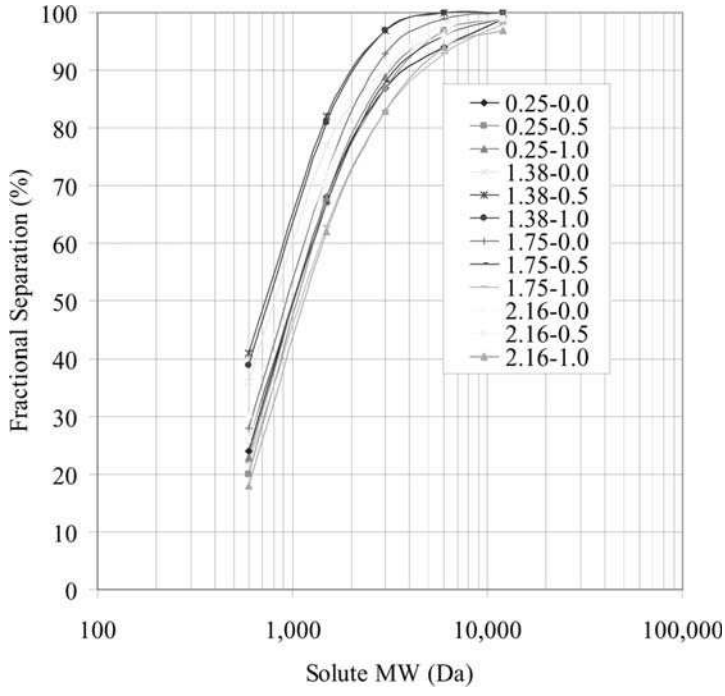

Figure 6. MWCO curves for twelve coupons cut from a 1 m wide by 2.5 m long membrane sheet. Legend code indicates position along length-position across width.

produced polyethersulfone membrane rated at 1 kDa MWCO were tested at 340 kPa: the intrinsic MWCO varied between 2.5 and 3.5 kDa, a very narrow range. The pure water flux averaged $124.3 \pm 16\,\mathrm{L \cdot m^{-2} \cdot h^{-1}}$, with a minimum/maximum of $77/167\,\mathrm{L \cdot m^{-2} \cdot h^{-1}}$. The observed MWCO is influenced by the flux variations, ranging from 2.5 to 10 kDa (not shown), which could impact the performance of these membranes if they were used to fractionate different species. Hence, the membrane's performance must take into consideration both the intrinsic separation and the actual flux.

CONCLUSIONS

An automated apparatus for characterization of ultrafiltration membranes using solute permeation has been described. The process lends itself well to automation due to the repetition of similar procedures. This avoids operator fatigue, chances of operator error, and essentially liberates the operator for other duties. Typical operator errors in manual characterization could be: data entries, sample collection timing, or insufficient permeate line flushing (sample contamination). The simultaneous characterization of

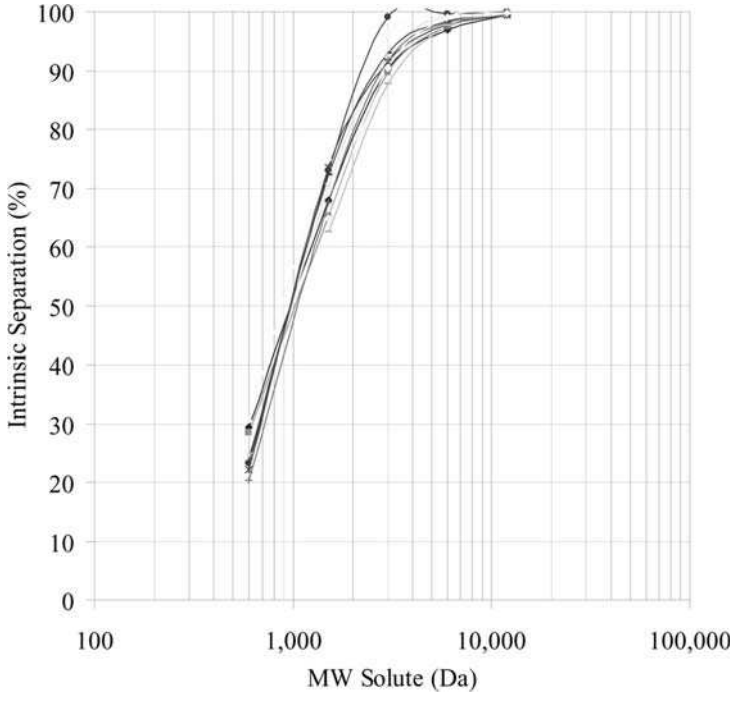

Figure 7. MWCO curves for a commercial polyethersulfone membrane rated at 1,000 Da MWCO.

12 samples gives more reliable information, allowing statistical analysis of the membrane uniformity and aids identifying random errors in the test procedure. A typical MWCO will take 24–36 hrs on the APU compared to 3–4 days on a manual system. Development of new membranes, quality control in production, and pre-selection of membranes for specific applications all benefit from the high throughput benchmark test.

The interpretation of the MWCO curve is in the hands of the operator, and dependent on the pressure (i.e., flux) and MW of the solutes, test cell design, and interpretive models. The APU is an apparatus to obtain this data with speed and accuracy.

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