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Experiments in Gas Permeation Membrane Processes*

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The area of gas stream separation and purification has been revolutionized in the past decade by the development of highly selective membrane systems. These new technologies are rapidly replacing antiquated ones and are being incorporated into many areas of chemical engineering. The chemical engineering curriculum can be improved by providing laboratory experience in gas permeation. This can be accomplished through the introduction of new laboratory experiments covering the fundamentals of membrane gas transport and the operation of a Prism® hollow-fiber membrane system. Experiments examine the basic concepts of membrane operation and the effects of process parameters on membrane performance. Experiments are conducted separating an air stream into purified nitrogen and enriched oxygen streams.

Nomenclature

D_i	diffusivity of component i in the membrane (L^2/t)
J_i	flux of component i (mol/L^2t, M/L^2t or L/t)
l	membrane thickness (L)
p_i	partial pressure of component i in the gas phase (M/Lt)
$\mathscr{P}_i$	permeability of component i (M/Lt)
Q	volumetric flow rate (L^3/t)
S_i	solubility of component i (mol/L^3 or M/L^3)
x_{im}	concentration of component i in the membrane (mol/L^3 or M/L^3)
x_i	concentration of component i in the gas phase (mol/L^3 or M/L^3)
α^*_{ij}	separation factor based on permeability differences (dimensionless)
α^i_j	selectivity of the membrane based on concentration differences (dimensionless)

Subscripts

1	permeate or permeate side of membrane
2	non-permeate or non-permeate side of membrane
i	component i
j	component j
p	permeate stream
f	feed stream.

INTRODUCTION

MEMBRANE technology is an interesting and technically exciting topic that deserves greater attention in the chemical engineering curriculum. Membrane processes show great promise for technical growth and wide-scale utilization. Therefore, effective integration of instructional material on membrane technology into the curriculum is warranted. This can be achieved through course and laboratory development at both the undergraduate and graduate level [1, 2].

It may not be possible within the highly-structured chemical engineering curriculum to add new courses or even modify existing courses to introduce adequately material on membrane processes. The chemical engineering laboratory provides the unique setting to introduce membrane principles, design and applications. Most schools have a senior level unit operations laboratory in which students typically perform structured or open-ended experiments. Most of these are bench-scale or pilot-scale experiments that give students a 'hands-on' experience. It is usually easier to add one or two new experiments to a laboratory than to change a course.

The National Science Foundation and industry have funded the development of laboratories at Manhattan College in the area of advanced separation processes, particularly membranes. We have successfully developed experiments in liquid separation membrane processes and have reported these results in the literature [3–9]. These papers discuss simple laboratory experiments that can be conducted using either bench-scale or pilot-scale membrane systems. Some of these systems were custom designed for the laboratory development projects. This paper describes experiments in gas permeation membrane processes.

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BACKGROUND

Membrane processes are unit operations utilized for liquid and gas stream separations. A membrane is an ultra-thin, semipermeable barrier separating two fluids that allows the selective transport of components from one fluid to the other. It is the selectivity of the membrane material that gives the process its utility and potential to separate a variety of process streams. The family of membrane processes includes the unit operations that are used for liquid stream separations; reverse osmosis, ultrafiltration, microfiltration, dialysis, electrodialysis, pervaporation and liquid membrane processes. Membrane processes used to separate gaseous feeds are comprised of those using porous and non-porous membranes.

Gas permeation is the term typically used to describe a membrane separation process using a non-porous semipermeable membrane. In this membrane process a gaseous feed stream is fractionated into permeate and non-permeate streams. The non-permeating stream is typically called the non-permeate in gas separation terminology and referred to as the retentate in liquid separations. Transport occurs by a solution–diffusion mechanism and membrane selectivity is based on the relative permeation rates of the components through the membrane. Each gaseous component transporting through the membrane has a characteristic permeation rate that is a function of its ability to dissolve in and diffuse through the membrane material. The mechanism for transport is based on solubilization and diffusion. The two chemical engineering relationships upon which the equations are based are Fick's law (diffusion) and Henry's law (solubility).

Diffusive flux through the membrane can be expressed by Fick's law related to the membrane system as

$$J_i = \frac{D_i}{l}(x_{im2} - x_{im1}) \quad (1)$$

where J_i is the flux of component i, D_i is the diffusivity of component i, l is the membrane thickness, x_{im2} is the concentration of component i inside the wall of the membrane on the feed side, and x_{im1} is the concentration of component i in the membrane on the permeate side.

Using Henry's law

$$x_{im} = S_i p_i \quad (2)$$

where S_i is the solubility constant for component i in the membrane and p_i is the partial pressure of component i in the gas phase.

Substituting Eq. (2) into Eq. (1) yields

$$J_i = \frac{D_i}{l}(S_i p_{i2} - S_i p_{i1}) \quad (3)$$

where p_{i2} and p_{i1} are the respective partial pressures of gas i on the feed and permeate side of the membrane. The permeation through the membrane is a function of solubility and diffusivity so let the permeability, $\mathscr{P}_i$, be represented by

$$\mathscr{P}_i = D_i S_i \quad (4)$$

Substitution of Eq. (4) into Eq. (3) gives the resulting relationship for the local flux through the membrane

$$J_i = \frac{\mathscr{P}_i}{l}(p_{i2} - p_{i1}) \quad (5)$$

Separation efficiency is based on the different rates of permeation of the gas components. A separation factor is frequently used to quantify the separation of a binary system of components i and j.

$$\alpha^*_{ij} = \frac{\mathscr{P}_i}{\mathscr{P}_j} \quad (6)$$

This is only one way to represent the relative degree of separation achieved. Another frequently used parameter relates the concentration of the feed and permeate streams. An apparent selectivity of the system can be described by

$$\alpha_j^i = \frac{x_{i1}/x_{j1}}{x_{i2}/x_{j2}} \quad (7)$$

where x_1 and x_2 represent the concentrations of components i and j in the permeate and feed streams, respectively.

Recovery in membrane operations is usually expressed as the ratio of the permeate flow rate to feed flow rate. For gas permeation systems a specific component recovery is usually calculated

$$\text{Recovery of component } i = \frac{Q_p x_{i1}}{Q_f x_{i2}} \quad (8)$$

where Q_p and Q_f represent the volumetric flow rates of permeate and feed streams, respectively.

The term stage cut or cut is used to define the ratio of the permeate flow to feed flow rate.

$$\text{Stage cut} = \frac{Q_p}{Q_f} \quad (9)$$

The use of membranes in gas separation was commercialized by Monsanto in the mid-1970s with the development of the hollow-fiber Prism® system [10]. Monsanto Company won the 1981 Kirkpatrick Chemical Engineering Achievement Award for development of the Prism® membrane system. Their hollow-fiber membrane concept allowed for the first time the practical use of membranes in large-scale gas separations. The technology represents a low-cost, simple option for gas purification and separation. Today several firms, e.g. UOP, Air Products and Chemicals, Dow, Du Pont and Grace, produce gas permeation membrane units.

Gas permeation systems are finding their way into both traditional and emerging engineering areas. Originally systems were developed for hydrogen recovery, but numerous applications are in use or in development. Applications are found in gas recovery for waste gas streams, landfill gases [11], ammonia [12] and petrochemical production [13, 14], etc. Gas permeation membrane systems are also utilized in gas generation and purification, e.g. nitrogen and enriched-oxygen, for chemical and petrochemical processing, food processing and storage, microelectronics manufacturing and the medical/health-care industry [15, 16].

EXPERIMENTAL SYSTEM

Recognizing the need to instruct undergraduate engineers in membrane gas separation, Permea, Inc. (St Louis, MO., USA), a Monsanto Company*, has developed a laboratory-scale version of its commercial membrane gas separation unit [17]. The unit was designed specifically for undergraduate engineering laboratories. Permea has built into their system price a significant discount. Engineering schools across the United States and several in Europe have purchased units, and Permea has worked with several chemical engineering departments to develop basic experimental methodology. Texas Tech University has produced several instructional manuals that accompany the system and has also produced a very detailed video tape on system operation [18, 19]. Their group has also devoted a significant amount of time to modelling the system and evaluating various process parameters. The Norwegian Institute of Technology has also been involved in the development project for Permea and has investigated several design aspects of the system [20]. Clements at the University of Nebraska has described basic membrane fundamentals and system design [21].

The system is designed for versatility in the study of variables such as flow rates and pressures. Prism® membrane modules used in the system are similar to industrial units. The instructional system is designed to operate easily and effectively in laboratory conditions. For example, it can separate air (21% O_2, 79% N_2) using normal laboratory pressures (100 p.s.i. or 6.895×10^5 Pa) and temperatures to produce a purified nitrogen stream and an oxygen-enriched stream.

The basic system consists of four hollow-fiber Prism® membrane modules which are mounted together on a common manifold. Each membrane module has approximately 2.7 m^2 of membrane area. The modules have an overall size of 1 in. (25.4 mm) diameter and are either 3 or 5 ft (0.91 or 1.52 m) long (depending upon the model) and are connected in series (the basic factory design). The Manhattan College system is composed of the 5 ft (1.52 m) long modules. The membranes are hollow fibers with diameters of approximately 500 μm. They are manufactured by a unique process and are largely composed of poly-sulfone post-treated with silicone (to eliminate defects).

The functioning of the laboratory-scale modules are identical to the larger ones used in commercial installations. Figure 1 shows how the hollow fiber configuration is used in air fractionation. The laboratory-scale system was designed by Permea to conduct experiments on air fractionation though other gases may be used if the proper design and safety requirements are met. A process flow diagram of the basic system used to conduct the experiments described here is presented in Fig. 2.

The compressed air stream to be separated is first sent to a series of coalescing filters and a pressure gauge. The dried air then enters the feed port at the first membrane module. Since the modules are connected in series (standard factory arrangement) the non-permeate from the first module becomes the feed to the second and so on. The non-permeating stream, which is the purified nitrogen, is passed through a flow meter and a measurement is made for its oxygen content with an oxygen analyzer. The standard system has a dry gas test meter that is quite accurate and we have

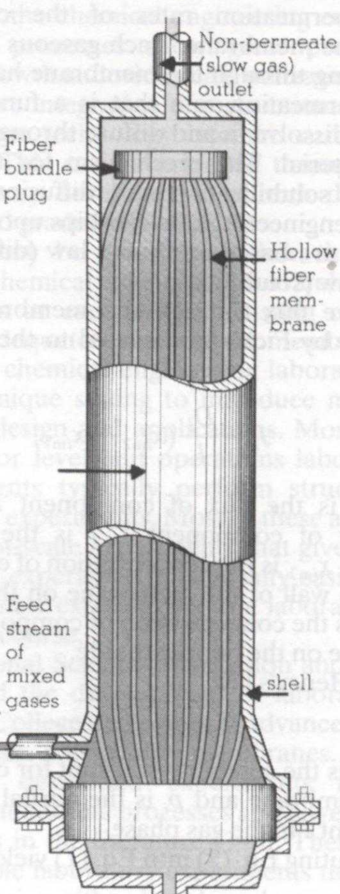

Fig. 1. Hollow-fiber membrane configuration as applied to air separation. (Courtesy: Permea, Inc.)

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* Permea is now owned by Air Products and Chemicals, Inc.

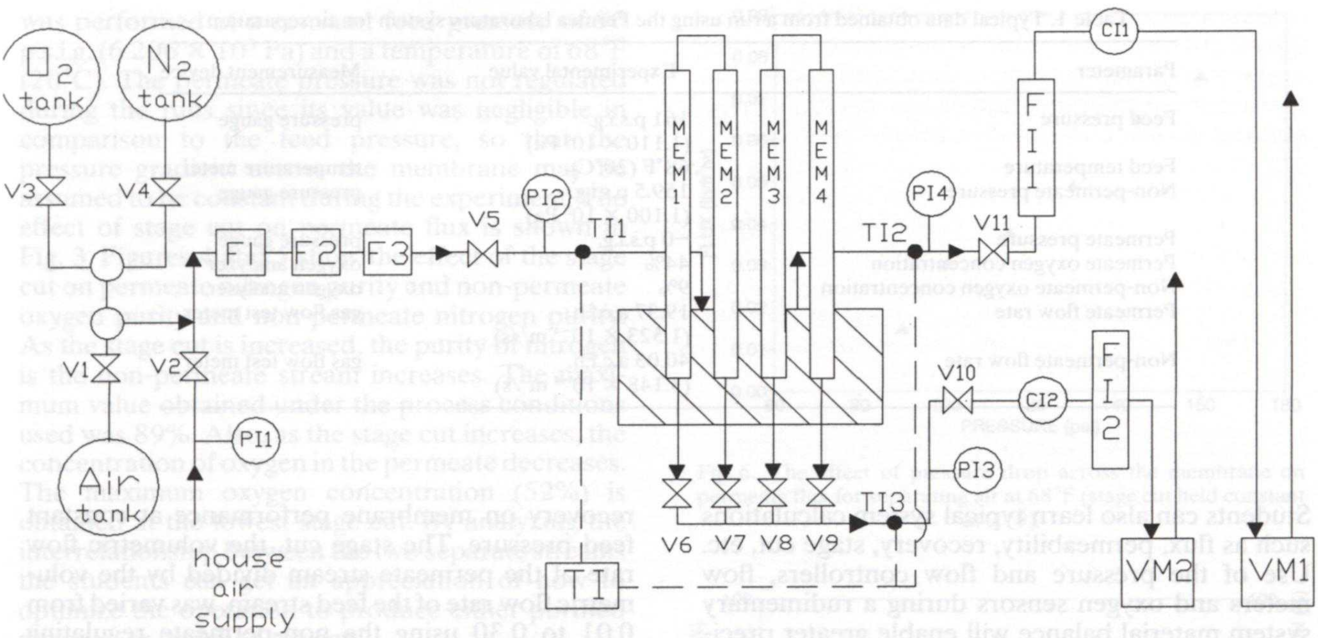

Fig. 2. The Permea laboratory system basic process flow arrangement (not to scale). Prefilter, F1; coalescing filters, F2, F3; feed pressure gauge, PI2; feed temperature thermocouple, TI1; temperature meter, TI; membrane modules, MEM1, MEM2, MEM3, MEM4; membrane module valves, V6, V7, V8, V9; non-permeate temperature thermocouple, TI2; non-permeate pressure gauge, PI2; non-permeate regulating valve, V11; non-permeate rotameter, FI1; non-permeate oxygen concentration meter, CI1; non-permeate volume meter, VM1; permeate temperature thermocouple, TI3; permeate pressure gauge, PI3; permeate regulating valve, V10; permeate oxygen concentration meter, CI2; permeate rotameter, FI2; permeate volume meter, VM2.

added a rotameter so that instantaneous flow rates can be observed. A needle valve is installed on the non-permeate line to adjust flow rate. A control valve can also be placed on the permeate stream to regulate the permeate-side, downstream pressure. Permeate flows from each module and is combined as a composite stream before being analyzed. Valves are placed at the permeate outlet of each module so individual modules can be isolated from the rest of the system. A flow meter and oxygen analyzer are also placed on the enriched-oxygen, permeate stream.

The system can be constructed to have all necessary piping to increase its ability to run different experiments and therefore ensure the long-range impact of the laboratory development. Control valves can be included so the membrane modules can be connected in a parallel or series–parallel arrangement. A sampling valve can be installed at each stage to study the multistage mass transfer. Additional prefilters are a good idea to protect the system from feed impurities found in laboratory air supplies and thereby increase the functional life of the system. In-stream oxygen analyzers are supplied with the standard Permea system. If experiments are planned for separation of gaseous mixtures other than air it is necessary to have a specific gas analyzer or a gas chromatograph.

The manufacturer recommends some limitations on the standard unit which should be followed unless the user has permission of Permea. Maximum feed air flow rate is 4 ft^3/min (1.89×10^{-3} m^3/s) and a pressure of 220 p.s.i.g. (1.517×10^6 Pa). Although one of the best features of the system is that experiments can be performed using a standard house air-supply system, most schools have compressors that do not go over 100 p.s.i.g. (6.895×10^5 Pa). To run experiments at higher pressures several options can be employed. To make the experimental system self-contained a separate compressor can be purchased with a sufficiently sized supply tank, or gas cylinders can be purchased for the experiments at high pressure. Both of these options are quite expensive. It is usually best to take the existing house air-supply compressor and modify it slightly to produce a smaller output of higher-pressure air. Concerns over use of the system outside of the intended temperature range and oxygen concentration are discussed below.

EXPERIMENTAL RESULTS

Several types of experiments are possible with the Permea laboratory system. A preliminary experiment can be done to check overall material and component balances for this separation process. This can be done at any experimental setting of the system and gives the students experience in performing calculations on a gaseous basis. Data from a typical run with the system are presented in Table 1. During this experiment, the students run the system at a given feed pressure and obtain data for the permeate and non-permeate flow rates and oxygen content. The material balances can be verified and the students can get a basic appreciation of the nature of separation being performed.

Table 1. Typical data obtained from a run using the Permea laboratory system for air separation

Parameter	Experimental value	Measurement device
Feed pressure	161 p.s.i.g. $(1.110 \times 10^6$ Pa)	pressure gauge
Feed temperature	68°F (20°C)	temperature meter
Non-permeate pressure	159.5 p.s.i.g. $(1.100 \times 10^6$ Pa)	pressure gauge
Permeate pressure	~0 p.s.i.g.	pressure gauge
Permeate oxygen concentration	44%	oxygen analyzer
Non-permeate oxygen concentration	9%	oxygen analyser
Permeate flow rate	19.37 a.c.f.h. $(1.523 \times 10^{-4}$ m^3/s)	gas flow test meter
Non-permeate flow rate	40.03 a.c.f.h. $(3.148 \times 10^{-4}$ m^3/s)	gas flow test meter

Students can also learn typical system calculations such as flux, permeability, recovery, stage cut, etc. Use of the pressure and flow controllers, flow meters and oxygen sensors during a rudimentary system material balance will enable greater precision for later process variable studies.

Typical experiments that can be conducted examine the effects of process variables on the separation efficiency [22]. The majority of these experiments are easily performed due to on-line analysis with the oxygen sensors and accurate gas flow meters. Two of the process variables that can be varied on the basic experimental set-up are pressure drop across the membrane and flow rate. By adjusting the feed pressure and maintaining the permeate pressure a pressure gradient can be obtained. Varying the non-permeate flow rate using a micrometer valve produces different feed flow rates and therefore different recoveries or stage cuts can be obtained. To generate some additional information about the system the individual modules can be isolated during the study. This allows a measurement of the variation between the individual modules in the overall system.

A good experiment using the system examines the effects of varying the stage cut or system recovery on membrane performance at constant feed pressure. The stage cut, the volumetric flow rate of the permeate stream divided by the volumetric flow rate of the feed stream, was varied from 0.01 to 0.30 using the non-permeate regulating valve. Typical plots showing the effect of this process variable on flux and process stream compositions are shown in Figs 3–5. The experiment

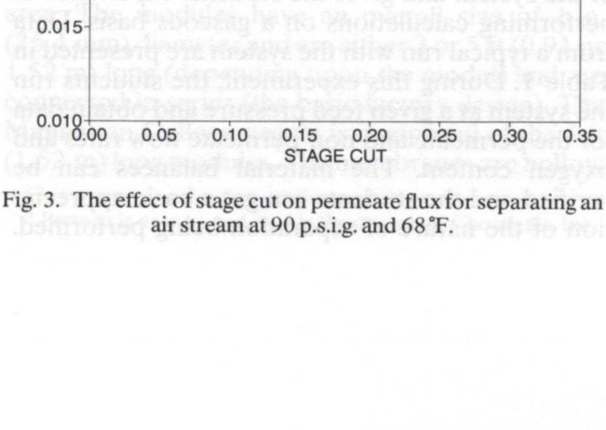

Fig. 3. The effect of stage cut on permeate flux for separating an air stream at 90 p.s.i.g. and 68°F.

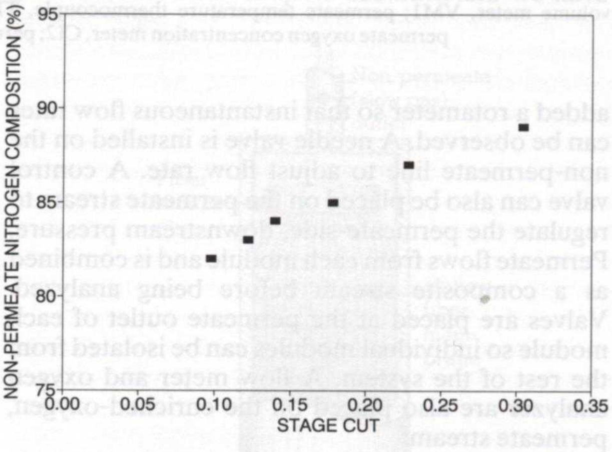

Fig. 4. The effect of stage cut on nitrogen concentration in the non-permeate for separating an air feed stream at 90 p.s.i.g. and 68°F.

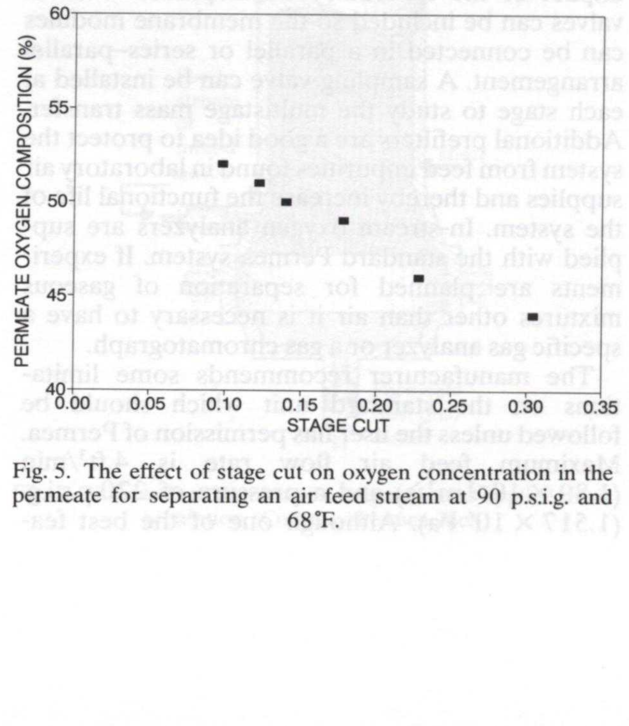

Fig. 5. The effect of stage cut on oxygen concentration in the permeate for separating an air feed stream at 90 p.s.i.g. and 68°F.

was performed at a constant feed pressure of 90 p.s.i.g. (6.206×10^5 Pa) and a temperature of 68°F (20°C). The permeate pressure was not regulated during the runs since its value was negligible in comparison to the feed pressure, so that the pressure gradient across the membrane may be assumed to be constant during the experiment. The effect of stage cut on permeate flux is shown in Fig. 3. Figures 4 and 5 show the effect of the stage cut on permeate nitrogen purity and non-permeate oxygen purity and non-permeate nitrogen purity. As the stage cut is increased, the purity of nitrogen is the non-permeate stream increases. The maximum value obtained under the process conditions used was 89%. Also, as the stage cut increases, the concentration of oxygen in the permeate decreases. The maximum oxygen concentration (52%) is obtained at the lowest stage cut. By analyzing the interrelationship between the two separate streams the students can get an appreciation of how to optimize the operation to produce either purified nitrogen or enriched oxygen.

Another typical experiment is to vary feed pressure at constant stage cut or recovery. In this series of runs the students can again examine the effect of a key process variable on membrane separation performance. Experimental correlations can be made between feed pressure and permeate flux, and permeate and non-permeate component concentrations. As the pressure increases the permeate flow rates increases, but stream compositions remain relatively constant (over the pressure range evaluated). Figures 6 and 7 show the effect of pressure on membrane separation performance.

Other more detailed experiments would involve system modifications. These experiments are not that difficult to perform, but do require some additions to the original basic configuration used in the studies previously described. Some of these experiments are summarized below.

The effect of feed temperature on membrane performance can be analyzed by preheating the feed. Check with Permea before performing this experiment since the temperature limit on the oxygen sensors supplied with the basic unit (Johnson & Johnson, Critikon Model 2000) is 40°C. This experiment demonstrates the Arrhenuis-type effect of temperature on the permeation rate. Permeate flux exponentially increases with temperature. It is possible to conduct an experiment and calculate the activation energy for separation.

Another experiment would examine the effect of feed concentration on membrane performance by varying the oxygen content in the feed stream. This can be done by using two gas cylinders containing pure nitrogen and oxygen and a manifold for blending. Caution should again be used before running this experiment. Although the Texas Tech group has worked with pure oxygen streams, Permea has warned our group to keep oxygen levels less than 50%. If other oxygen concentrations or other gases are planned to be used, contact Permea to discuss safe operating limits.

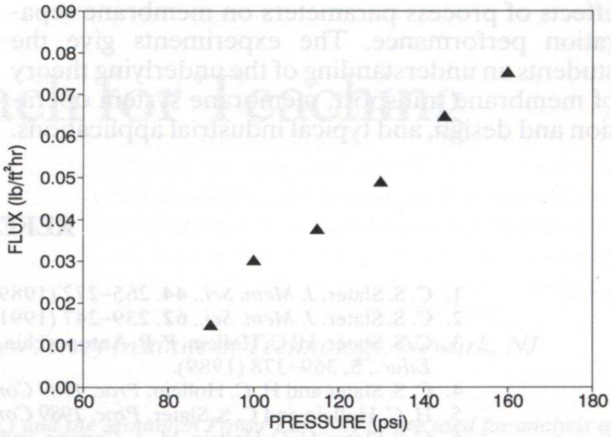

Fig. 6. The effect of pressure drop across the membrane on permeate flux for separating air at 68°F (stage cut held constant at 0.19).

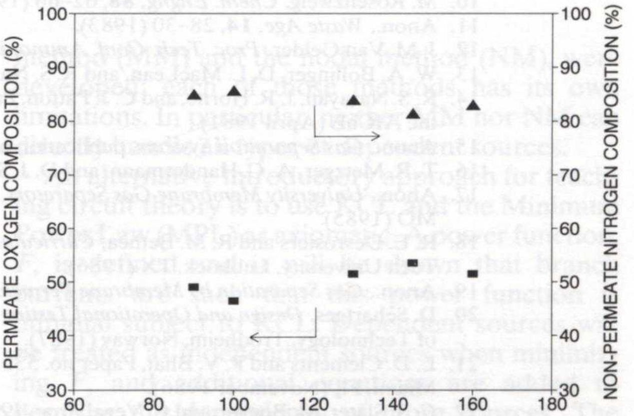

Fig. 7. The effect of pressure drop across the membrane on non-permeate nitrogen composition (triangles, right ordinate) and permeate oxygen composition (squares, left ordinate) for separating air at 68°F (stage cut held constant at 0.19).

One of the more popular design studies is to modify the simple series arrangement of the system. This does require some work in adding additional pipe and valve arrangements. Both the Norwegian Institute of Technology manual [20] and the paper by Clements of the University of Nebraska [21] describe the different configurations possible. These are variations of the series, parallel and series–parallel system design layouts. This experiment is good in showing the design layout patterns used in large-scale systems. A parallel feed system accommodates higher throughputs and a series system produces higher recoveries.

SUMMARY

Students can gain exposure to the rapidly growing field of membrane technology by performing experiments in gas permeation. A laboratory gas permeation system manufactured by Permea, Inc., a Monsanto Company, was used to analyze the effects of process parameters on membrane separation performance. The experiments give the students an understanding of the underlying theory of membrane transport, membrane system operation and design, and typical industrial applications.

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