

DEVELOPMENT OF INORGANIC MEMBRANES FOR GAS SEPARATION

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Manuscript

Introduction

In previous years, the annual report concentrated on the work that was performed in the immediate past year. This year, in order to emphasize the great amount of progress that has been made, we will summarize our process and present the many membranes that we fabricated with high separation factors.

One of our past major problems has been the classified status of much of our fabrication process. DOE has developed a review process whereby developed membranes can be reviewed for classified information and non-proliferation risk. When a membrane passes that review process the membrane can be declared unclassified and available for commercial manufacture. Currently, we have fifteen membranes/filters approved for commercial manufacture. At present, Pall Corporation is the only company licensed to manufacture these membranes/filters.

Membrane Fabrication

If an inorganic membrane is homogeneous, then its permeance is inversely proportional to the membrane thickness. For gas separations, the effective mean pore diameter should be 2 nm or less. With such small pores, the membrane must be very thin, a couple of microns and preferably less. In order for such a thin membrane to be useful, it must be layered on a strong porous support material, either metal or ceramic. Metal is preferably because modules can be more easily and less expensively constructed. The primary membrane layer can be applied directly to the support material (preferably a tube with the layer on the inside of the tube for protection of the layer). An intermediate layer with an intermediate pore size can be applied to the support tube first to help produce a thinner and more uniform primary membrane layer. The primary layer should have a mean effective pore diameter of 10 nm or less and preferably as small as 2 nm. Once the primary layer is in place various chemical treatments are used to reduce the effective pore diameter to the desired value, as low as 0.5 nm.

It is extremely difficult to fabricate a membrane with zero defects. We have several different methods to reduce the effective pore diameter of a defect or eliminate the defect. Elimination of defects is critical because a disproportionate amount of flow will be through the defect and that can seriously reduce the magnitude of the membrane separation factor. We have a measurement that can be used with an in-house developed model¹ to estimate the percentage of flow through the defects and the amount the separation factor will be lowered.

The Importance of Membrane Characterization and Testing

The two most important characteristics of inorganic membranes are the permeance and the ideal separation factor. The permeance is a measure of the gas flow rate per unit area per unit pressure difference. The unit that we use is standard cubic centimeters per minute per square centimeter per centimeter of mercury, abbreviated as sccm/sq cm/cm Hg. A more fundamental unit is permeability, which is the permeance multiplied by the thickness of the membrane. In most cases, we don't know the thickness of the membrane very accurately and so permeance is a more practical unit.

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The ideal separation factor is meaningful only with respect to a binary mixture of two gases. It is the ratio of the permeance of the two gases measured at zero pressure where there is no interaction or momentum exchange between the two gases. Each gas flows through the membrane as if the other gas was not there. The ideal separation factor for a given temperature can be estimated by measuring the permeance of each gas as a function of average pressure and extrapolating the permeance to zero average pressure. The ideal separation factor is then the ratio of the zero pressure permeances.

The Different transport mechanisms behave differently as the pore diameter is reduced. They also behave differently with respect to temperature at different pore diameters. Measuring pore diameters less than 2 nm is extremely difficult. It is critically important to be able to follow the changes in the transport mechanisms of different gases to help understand where you are as the pore diameter is reduced. We have several in-house developed theoretically based models to help understand the transport mechanisms. We have also developed a detailed and involved protocol that we use to follow those changes. The most important model which has been developed is the Hard Sphere Model²⁻³, which has been reported previously. It basically takes into account the size of the gas molecule. The diameter of a pore appears to the molecule to be the pore diameter minus the diameter of the molecule (or its equivalent hard sphere). Without taking into account the molecular diameter, the separation factor for free molecule diffusion (Knudsen flow) is the square root of the molecular weight ratio. With the molecular diameter consideration, the separation factor for free molecule diffusion (Knudsen flow) is the square root of the molecular weight ratio (Knudsen separation factor) multiplied by the cube of the ratio of the difference between the pore diameter and the molecular diameter for each molecule. The variation in the separation factor with pore diameter for several gas pairs is shown in Figure 1. For this transport mechanism, the model gives an interpolation formula between the Knudsen separation factor and molecular sieve separation factor. When the pore diameter becomes equal to or less than the larger of the two molecules, the larger molecule cannot pass through the membrane and the separation factor becomes infinite for a molecular sieve. As can be seen in Figure 1, the larger the difference in the molecular diameters the larger the pore diameter when the separation factor becomes infinite. This clearly shows the potential for achieving very large separation factors, even at pore diameters larger than the molecular sieve pore diameter, when there is a difference in the molecular diameters of the gas pair.

Of course, free molecule diffusion is not the only transport mechanism. The next most important transport mechanism is surface flow. Surface flow occurs when there is significant adsorption of a gas on the walls of the membrane. While the molecules are adsorbed on the membrane surfaces, they are in motion and can diffuse on the surface. In general, the heavier the molecule or the larger the interaction potential between the membrane surface and the molecule, the larger the adsorption and the more surface flow. Since this transport mechanism favors the heavier molecule, it tends to decrease the separation factor. Surface flow has been included in the full mathematical transport model³. However, adsorption and surface flow measurements are required to evaluate constants in the mathematical formulation. Only one attempt has been made to date. Carbon dioxide adsorption and appropriate permeance measurements were made at 25 °C with an alumina membrane. Model calculations were then made for the binary pair, helium and carbon dioxide. Zero surface flow for helium was assumed. The results are shown in Figure 2. As the pore diameter decreases, the gas phase diffusion decreases and the surface flow increases primarily because the relative amount of surface area increases relative to the pore volume. This causes the separation factor to decrease until the pore diameter approaches the diameter of the carbon dioxide and the transport of the carbon dioxide decreases sharply and the separation factor increases sharply. This calculation was based on the flow of the individual pure gases. It doesn't take into account the fact that the adsorption of the carbon dioxide decreases the size of the pore diameter for the helium. Therefore, in mixed gas separation, the separation factor will be even smaller than is shown in Figure 2. It should be pointed out that the separation factor drops below unity and becomes less than one. Which means that the carbon dioxide permeance is larger than the helium permeance. Below we will show some actual permeance measurements where the carbon dioxide permeance is larger than the helium permeance.

Permeance Measurements

Rapid and highly accurate permeance measurements are the heart and soul of our membrane development management protocol. Single point permeance measurements are of little value. Permeance is measured as a function of average pressure. A

linear regression of permeance vs. average pressure (we use the sum of the feed pressure and permeate pressure, twice the average pressure, and refer to it as sigma P or pressure summation) provides valuable information. Our standard test membranes are tubes about 1 cm in diameter and 20 to 25 cm long. Initial testing is performed with air at room temperature. A series of 5 to 25 permeance measurements is made over an average pressure range from about 50 to 200 cm Hg. A linear regression is calculated and the zero permeance, a permeance deviation factor, and the permeance at an average pressure of 75 cm Hg is

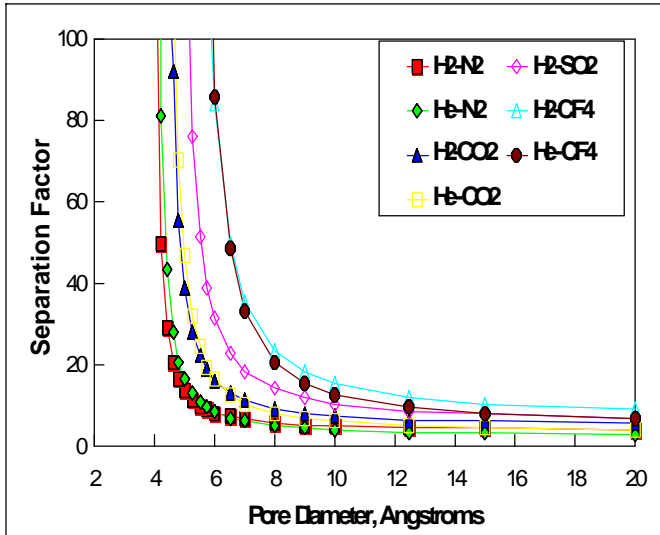


Figure 1 Separation factors for gas pairs with different relative sizes as a function of pore diameter using the Hard Sphere Transport Model.

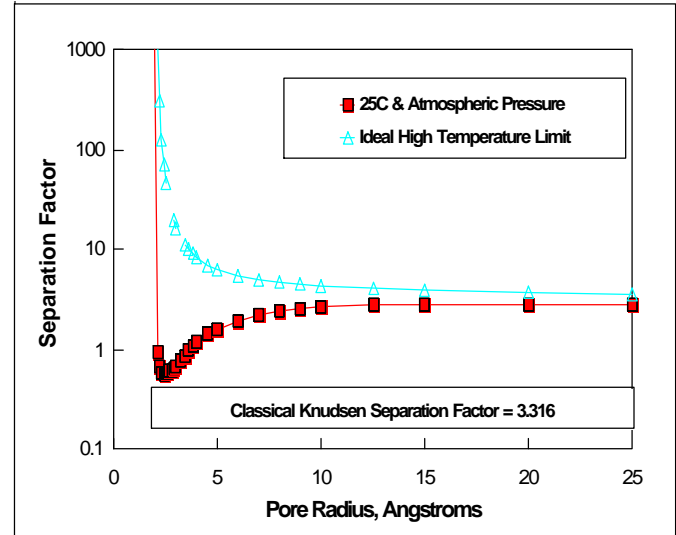


Figure 2 He-CO₂ separation factors at 25 °C calculated from the Full Transport Model compared with the Hard Sphere Free Molecule Diffusion Model

reported. The permeance deviation factor is the ratio of the slope of the linear regression to the zero pressure permeance. A positive value may indicate viscous flow from defects in the membrane. The measurements usually take less than ten minutes. These measurements are made on the membrane at every stage of development. The permeance data is measured on the support tube and at every addition in the fabrication of that membrane. For any given membrane development, 10 to 100 similar membrane preparations will be followed. If at any stage, a membrane differs significantly from expectation, it will be discarded. It is not unusual to make 1000 or more of these measurements during a year.

Membranes that show particular promise or membranes that are being used to monitor the progress of fabrication steps go to the next level of permeance testing. At this level, permeance measurements are made over the same average pressure range and at three temperatures, 25 °, 150 ° and 250 °C. This series of measurements is made with three or four pure gases selected from helium, hydrogen, oxygen, argon, carbon dioxide, carbon tetrafluoride, and sulfur hexafluoride. A linear regression with pressure summation (sum of feed and permeate pressure) is made at each temperature and for each gas. The ideal separation factor for each gas with respect to helium is calculated using the zero pressure permeances. For each gas, a linear regression of the natural logarithm of the ideal separation factor is calculated with respect to the inverse of the absolute temperature. Surprisingly, this relationship does usually result in a linear relationship. This relationship can then be used to estimate the ideal separation factor at any temperature. More important, the intercept at $1/T=0$ provides a significant parameter. At infinite temperature ($1/T=0$), no adsorption would be expected. Therefore, the flow is primarily free molecule diffusion. The equation used to calculate the results in Figure 1 can be used with the ideal separation factor at $1/T=0$ and the molecular diameters to calculate a mean pore diameter for the membrane. While the accuracy of this pore diameter calculation is unknown, it does provide a parameter to track the progress in reducing the membrane pore diameter.

The variation in permeance with pressure summation is shown in Figures 3 and 4 for a membrane with minor but significant defects. For pure free molecule diffusion, the permeance should be constant and independent of pressure. The positive slopes of the linear regressions for the four gases indicate some viscous flow. For viscous flow, the slopes are proportional to the product of the viscosity and the molecular velocity. When the slopes are not proportional to the product of the viscosity and the molecular velocity then the behavior is due to some other transport mechanism. The permeance is proportional to the ratio of the molecular velocity to the absolute temperature. If the permeance is multiplied by the inverse of the ratio then for free molecule diffusion, the intercept should be the same for all gases. Or perhaps a better way to look at the problem is the ideal separation factor for a gas pair is the ratio of the zero pressure intercepts. For free molecule diffusion (Knudsen flow), the ideal separation factor is the square root of the molecular weight ratio. Deviations from square root of the molecular weight ratio ideal separation factors indicate that other transport mechanisms are involved. An example of significantly different intercepts is

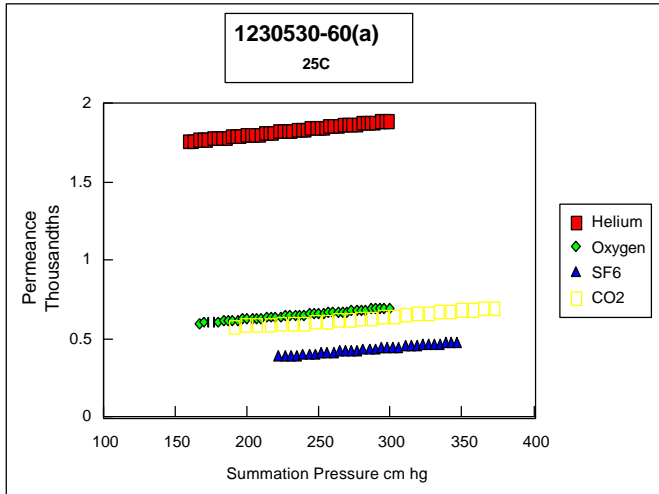


Figure 3 Variation in permeance with pressure at 25 °C for a membrane with minor defects.

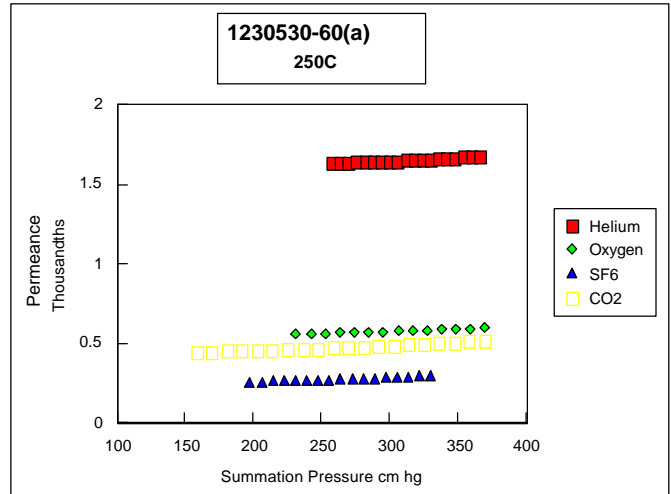


Figure 4 Variation in permeance with pressure at 250 °C for a membrane with minor defects.

shown in Figures 5 and 6. With this membrane the permeance of carbon dioxide at 25 °C is about 2.5 times larger than the permeance of helium. At 250 °C the carbon dioxide permeance is less than one half the permeance of helium. This indicates a high degree of surface flow with this membrane. In addition, the linear regression shows a negative slope. This likely indicates that the adsorption of carbon dioxide is large enough that it reduces the pore size of the membrane as the pressure increases. This implies that, in a gas pair mixture with helium, the adsorption of carbon dioxide would also reduce the pore size for helium and reduce the helium permeance to a value less than what it was for the pure gas. This would suggest that the separation factor for carbon dioxide would be even higher than indicated by the individual pure gas permeance. There is a surface flow component for all of the gases shown except for helium. The surface flow component shows up most prominently in the zero pressure permeance. That is, apparently the surface component depends more on the relative amount of time the adsorbed molecules spend on the surface rather than the surface population. Not much surface flow is evident at 250 °C. The ratio of the permeance of carbon dioxide at 25 °C is more than 4 times the permeance at 250 °C. That is also true for sulfur hexafluoride. It should be noted that the permeance of helium increases with temperature, whereas the other gases decrease with increasing temperature. The variation of the permeance with pressure is essentially constant at 250 °C. At 250 °C, the ratio of the permeances (ideal separation factors) for helium and carbon dioxide is only about 2.3 indicating some remaining surface flow of carbon dioxide.

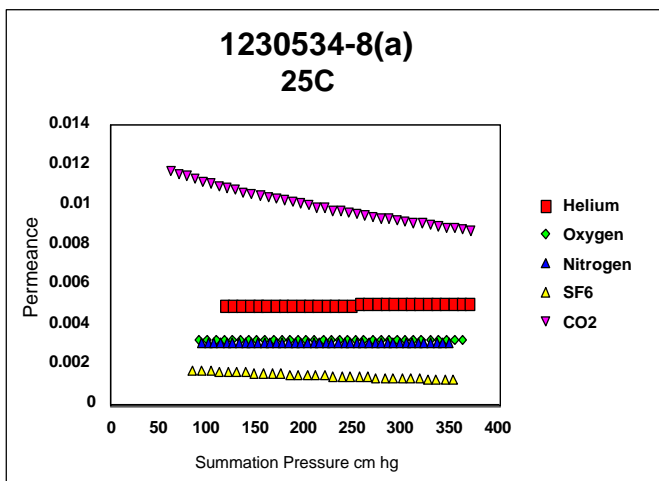


Figure 5 Variation in permeance with pressure at 25 °C for a defect-free membrane with a large surface flow component for some of the five gases.

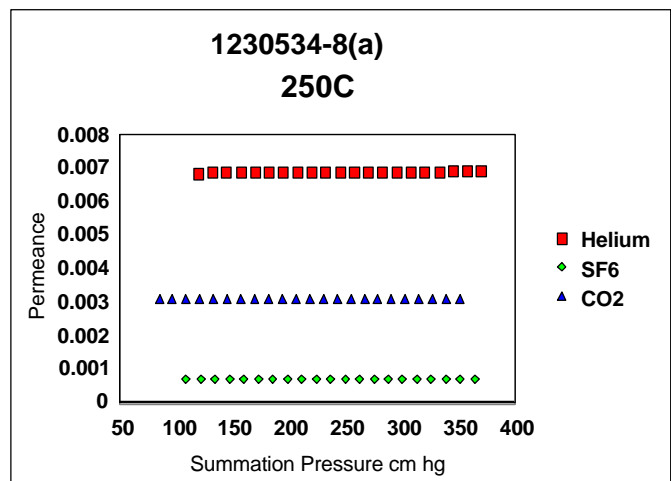


Figure 6 Variation in permeance with pressure at 250 °C for a defect-free membrane showing the decrease in surface flow with increase in temperature.

With the acquired permeance data, the ideal separation factors for the gas pairs are calculated from the permeance ratios. The natural logarithm of the ideal separation factor is plotted as a function of the inverse absolute temperature. A linear regression is performed and the intercept at $1/T=0$ is used with the molecular diameters of the gases to calculate a model estimated pore diameter as outlined in the protocol given above. The data shown in Figure 7 are used to make the calculation for the graphs shown in Figure 8 with the data obtained shown in the table below Figure 8. The data are surprisingly good considering the simplicity of the model and only three data points are available for each regression analysis. This is only a small sampling of the many membranes that we have evaluated. As can be seen readily, there is a great deal of information that can be acquired from individual pure gas permeance data when a sufficient amount of precise data is acquired over a range of pressures and temperatures.

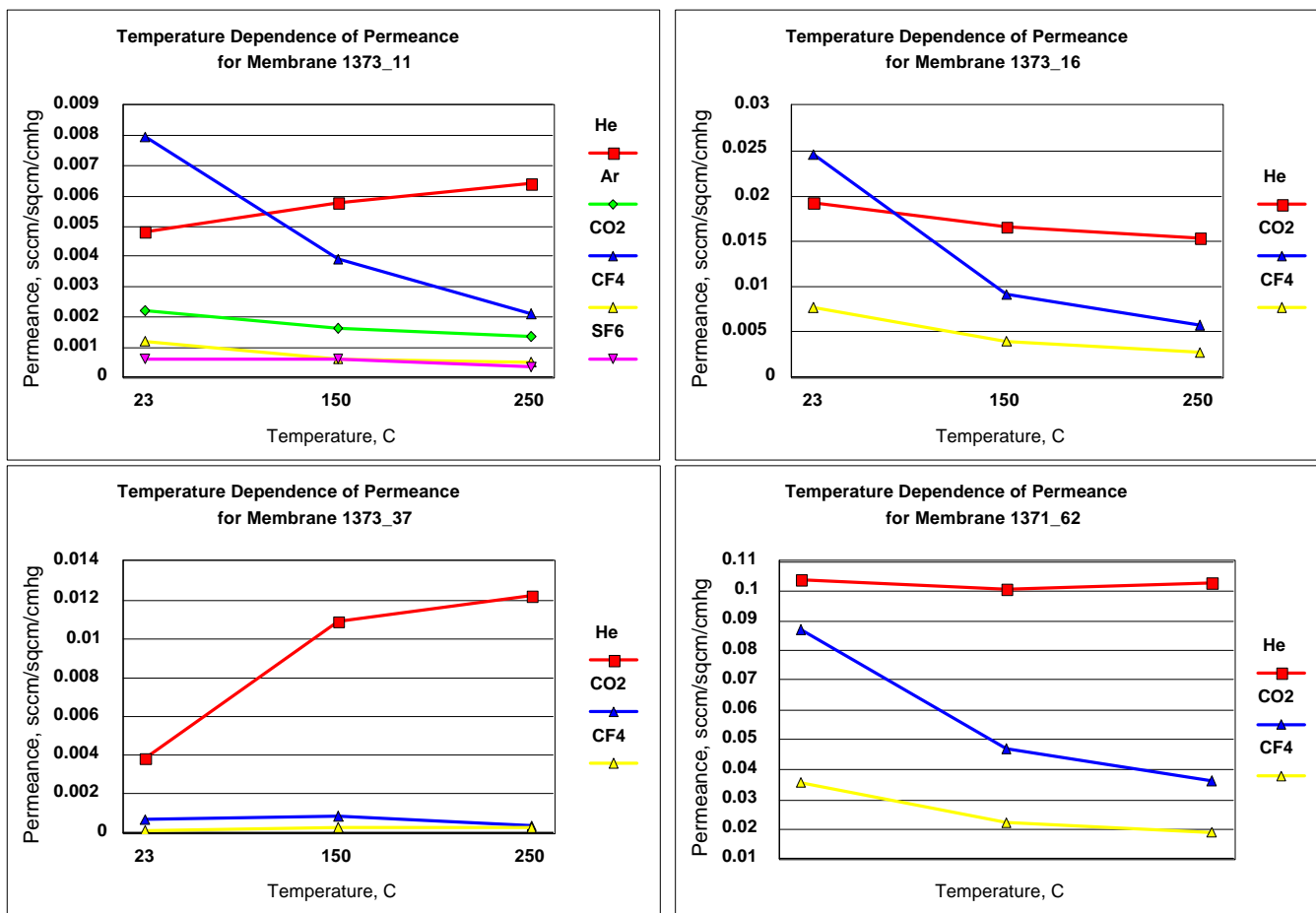


Figure 7 The temperature dependence of the permeance of four different membranes with helium, carbon dioxide, and carbon tetrafluoride, and with the addition of argon and sulfur hexafluoride on the top

For Ideal Free Molecule Diffusion (Knudsen Flow) the ratios of the permeances should be

- He/CO₂ = 3.316**
- He/CF₄ = 4.689**
- He/SF₆ = 6.041**
- He/Ar = 3.159**

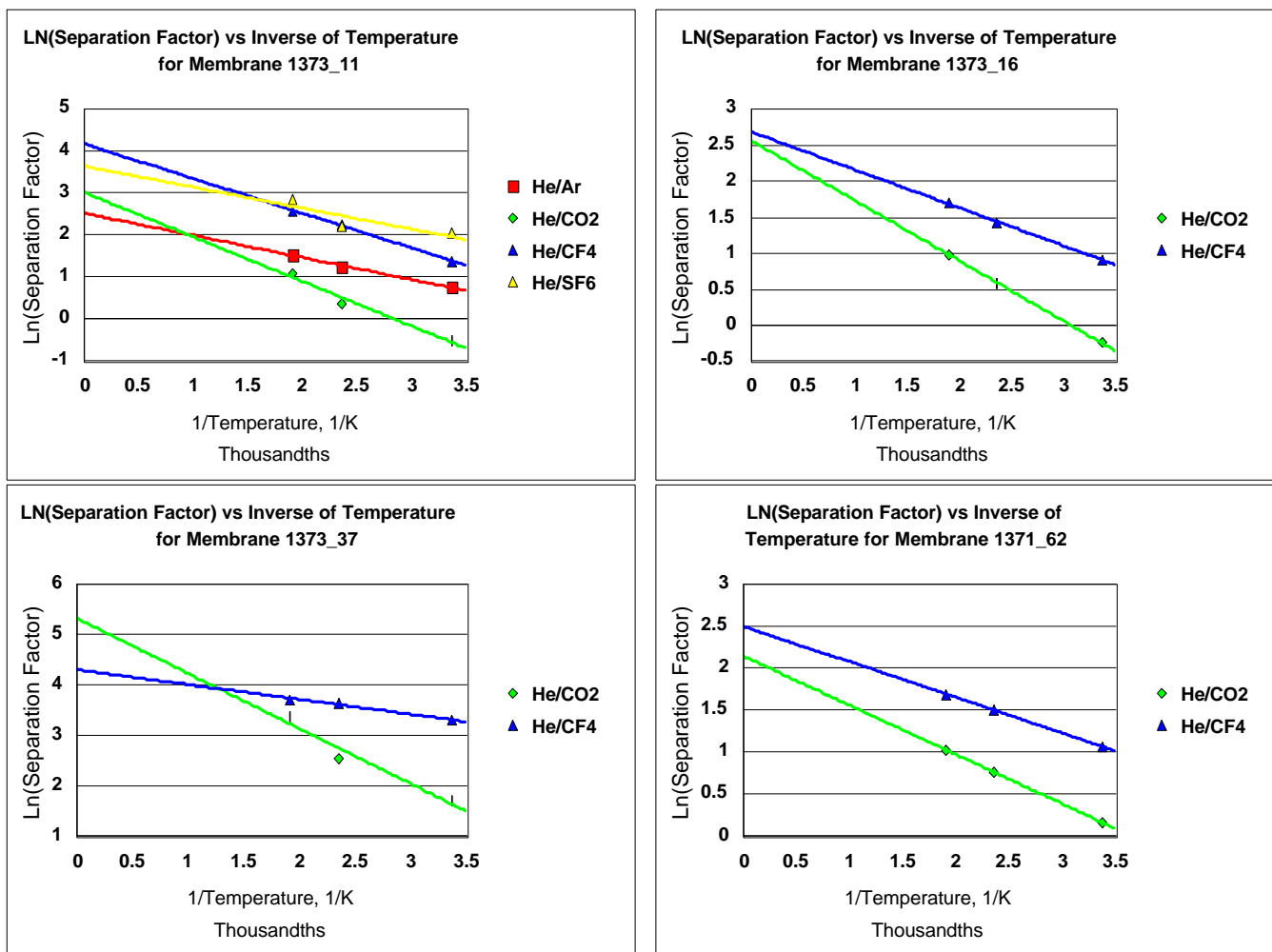


Figure 8. These graphs show the linear extrapolation of the natural log of the separation factor vs. the inverse absolute temperature.

Membrane	Gas Pair	Intercept	Separation Factor at 1/T=0	Model Estimated Pore Diameter, Angstroms
1373-11	He/Ar	2.53	12.6	4.33
	He/CO ₂	3.03	20.8	5.13
	He/CF ₄	4.19	65.8	5.70
	He/SF ₄	3.65	38.6	7.46
1373-16	He/CO ₂	2.56	13.1	5.52
	He/CF ₄	2.69	14.8	7.07
1373-37	He/CO ₂	5.34	208.7	4.36
	He/CF ₄	4.32	75.1	7.07
1371-62	He/CO ₂	2.15	8.61	6.09
	He/SF ₄	2.51	12.3	7.41

Mixed Gas Separation Measurements

The ideal separation factor calculated from zero pressure permeance values represents the highest separation that can be achieved with a given gas pair with the membrane used for the permeance measurements when there is no interaction or momentum exchange between the gas pair molecules. We have already discussed how adsorption of one of the molecules can change the pore size and thus the separation factor. As a gas pair travels through a membrane, the different species can collide with each other and transfer momentum. R. D. Present and A. J. deBethune have derived a method for calculating this momentum exchange⁵. To the first order, the momentum exchange depends on the pressure difference across the membrane.

In an operating separation system, a feed gas mixture is passed by a membrane and a portion of the gas, the permeate, passes through the walls of the membrane and a portion, the retentate, exits the membrane at the opposite end of the membrane. The fraction of the feed gas passing through the walls of the membrane, as the permeate, is enriched in the gas with the higher permeance. The retentate is depleted in the gas with the higher permeance. The ratio of the permeate flow rate to the feed flow rate is usually called the **cut**. The major variables in an operating gas separation system are the pressure ratio, the pressure difference across the membrane, the cut, and the temperature. All four of these variables effect the operating separation factor in a different way. In an operating gas separation system, the actual separation factor is strongly dependent on these operating conditions. For example, since the higher permeance gas is being withdrawn from the feed gas preferentially, concentration polarization will develop near the membrane surface. The geometry of the feed flow path in front of the membrane is important to minimize the effect of concentration polarization. In general, the separation factor measured in an operating separation system will be significantly less than the ideal separation factor.

IMTL has a State-of-the-Art, one of a kind, mixed gas separation system. The system is essentially a small single stage gas separation system. The membranes used are our standard 25 centimeters long, 1 centimeter diameter tubes. The feed gas enters one end of the tube, a fraction of the gas passes through the walls of the tube, and the remaining fraction passes out of the tube at the opposite end of the tube. The system can be operated at feed pressures up to 500 cm Hg, permeate pressures as low as 30 cm Hg, cuts between about 0.1 and 0.5 (or higher), and temperatures from ambient to 600 °C. However, only recently we have been able to operate up to 500 °C because of difficulty in sealing the membranes at temperature higher than 300 °C. Feed, permeate, and retentate mole fractions are measured with gas chromatography. Pressures are measured with high precision capacitance transducers and temperatures are measured with high precision thermistors. A flow meter is available to measure feed flow rates, but it is not a critical measurement to evaluate the separation factor. The cut is a critical measurement, but it can be calculated from the mole fraction of the three streams, especially if only binary mixtures are used for the measurements.

The beauty of the system is that we can acquire data to calculate actual separation factors over the operating range of the system. With the properly acquired data, we can extrapolate the measured separation factors to the ideal separation factor for that gas pair. **We have, in fact accomplished that with two different membranes.** While the amount of data available was not as much as desired, the extrapolated ideal separation factor from the binary mixed gas separation was within experimental error the same value as the ideal separation factor calculated from the pure gas permeance measurements. **I believe that is the first time such agreement has been made experimentally.**

Measurements such as these are necessary to understand how different membranes will operate in a real gas separation system. Once there is some basic understanding of the functioning of different types of membranes with various binary mixtures, the ideal separation factors (referenced to the same common gas) can be used to estimate the results that will be obtained with multi-gas mixtures.

Summary of the Highest Ideal Separation Factors Calculated from Permeance Ratios

Over the more than ten years of development work in inorganic membranes for hydrogen separation, we have tested thousands of membranes. Most of the work was directed toward obtaining data to help understand the transport mechanism of different gases at different temperature, pressures, and pore sizes. The data was essential in the development of the models and evaluations methods. Over that period, we have made a dozen membranes with significantly high ideal separation factors. Most years that was none, one, or two to report and none that could be released for independent corroboration. This is the first time we have compiled a list of the best membranes that have been evaluated and, in order to get this report ready, it is not fully

complete. The list does show that a great deal of progress toward achieving very high separation factors has been made. The data are consistent with the models we have developed.

Inspecting this data, it can be seen that surface flow of the heavier molecule plays a major role in reducing the value of the ideal separation factor at the lower temperatures. Surface flow becomes a larger factor the smaller the mean pore diameter. For the gases that we have used, surface flow is essentially negligible for temperatures of 250 °C and higher. This remains to be seen for other gases. For a given membrane and at temperatures of 250 °C and higher, the ideal separation factor increases with the increasing difference in molecule size between the two gases used in a binary mixture. It has also been observed that the permeance of helium and hydrogen increases with increasing temperature. More importantly, the rate of increase in helium permeance with temperature increases with decrease in pore diameter. This effect is not expected classically, but it is fortunate since that effect contributes significantly to higher ideal separation factors.

Listing of Membranes with Ideal High Separation Factors									
Separation Factor at 250 C and Extrapolated Value for 600 C									
Separation Factors (1)									
Gas Pairs									
	25 C	250 C	600 C	250 C	600 C	250 C	600 C	250 C	600 C
Sample #	Permeance	He/CO2	He/CO2	He/CF4	He/CF4	He/SF6	He/SF6	He/Ar	He/Ar
1371-37b	0.0015	25.7	64.6	42.4	70.0				
182686	0.00037	24.1		75.0					
1373-37	0.005	20.3	44.7	46.7	59.6				
1373-38a	0.0026	10.0				165.9			
1373-19	0.05	4.1		3.0	9.3	109.0		6.0	9.8
1373-19c		7.6				26.1			
1373-11	0.005	2.7		16.4		29.0		5.4	
1373-62b	0.013	5.0		10.3					
1373-97		2.7		10.3				4.4	
1373-62d						18.4			
1373-43	0.025	4.5				9.5			
1373-94b	0.01	4.3		8.4		10.9			
(1) Separation factors calculated from ratios of zero pressure permeance									
(2) Permeance is in units of std cc/min/sq cm/cm hg									
Knudsen Separation Factors									
	He	CO2		CF4		SF6		Ar	
Mole Wt	4.0026	44.0098		88.0046		146.01		39.948	
Kn Sep Fctr	Basis	3.316		4.689		6.040		3.159	
Diameter, A	2.56	3.99		4.70		5.51		3.42	

Conclusions

Much of the work in recent years has been toward increasing the membrane permeance, achieving repeatability with defect-free membranes, and using materials and techniques that can be approved by the DOE review process and manufactured on a large scale. Significant progress has been made in all these areas. We have significantly expanded our understanding of gas transport in inorganic membranes. We have significantly expanded our capability for fabricating inorganic membranes. We have developed and expanded our laboratory. We have a laboratory with extraordinary capability for characterizing and measuring the performance of inorganic membranes.

Acknowledgments

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