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REVIEW

Membrane Gas Separation: A Critical Overview

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INTRODUCTION

Membrane gas separation was one of the earliest applications of microporous membranes, as described originally by Thomas Graham in 1866 (1). However, despite the inherent and desirable advantages of this type of membrane toward high permeability, most of the early work on synthetic membrane gas separations has been carried out with the so-called homogeneous or dense membranes which are not necessarily asymmetric. The latter type of membrane is distinct from the purely homogeneous or nonporous and chemically modified membranes, viz., the vinylidene fluoride and other commercial films reported by Heyd and McCandless (2). The obvious reason for the lack of interest in microporous membranes, which is apparently responsible for their failure to achieve commercial success until a few years ago, is the empirical view that permeability and "permselectivity" are somewhat mutually exclusive or inversely related to each other. It is interesting to note that while virtually all the important engineering and technological concepts were well developed (see, for example, Kim and Kammermeyer (3), Stern and Walawender (4), Pan and Hab-

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Habgood (5), Hwang et al. (6), Sirkar (7) and Schulz et al. (8)), commercial realization came only in the late 1960s (9).

A number of workers studied the separation of gaseous mixtures with a variety of membranes and also from a fundamental standpoint. Attempts have been made, in some cases, to predict the membrane performance on the basis of some models. There has been, however, a lack of efforts toward a comprehensive model which takes into account the different modes of transport. Graham's work was considered to be based on Knudsen diffusion, whereas the existence and significance of other types of flow, notably surface flow, are well documented both for nonpolymeric (10, 11) and synthetic polymeric membranes (12). Based on extensive work on microporous carbon membranes, Ash et al. (13) stated that "flow of adsorbable gases through microporous membranes is often much enhanced by an extra component associated with mobile adsorbed films in concentration gradients. The total flows can then be very selective and can lead to good separation of mixtures." This is equally valid for porous synthetic membranes (14), and this particular mechanistic aspect is playing an increasingly important role in the development of inorganic membranes for gas separations.

TYPES OF GAS SEPARATION MEMBRANES AND SYSTEMS

In a comprehensive review on the growth of membrane technology, Lonsdale (15) described briefly the development of commercial membranes for gas separations. These membranes are basically of two types, viz., asymmetric and composite. Asymmetric membranes are generally obtained by drying Loeb–Sourirajan-type membranes, and they are considered to be composed of a dense and homogeneous skin layer supported by a porous backing of the same material. On the other hand, composite membranes are formed by depositing a thin and homogeneous layer or coating of a highly permeable polymeric material on a porous membrane of a different kind. In either case, the mechanism of transport affecting separation of a mixture of gases is almost invariably regarded as one of the solution–diffusion type, while the porous membranes lend themselves to different types of flow, namely, pore flow as well as surface flow. However, synthetic membranes of the latter type have received very little practical consideration, and apart from the early work of Sourirajan (16) and of Agrawal and Sourirajan (14, 17), the only reported work with such a membrane of cellulose acetate along with some silicone rubber membranes has been due, until recently, to Ohno et al. (18) who studied the separation of radioactive rare gases in mixtures of nitrogen–krypton and

helium–krypton. It is to be noted that the above porous membranes are generally asymmetric and these are not necessarily the same as those referred to as asymmetric dense or asymmetric homogeneous membranes.

Sirkar (19) gave a gross picture of pore size variation from the skin top through the transition layer to the top of the microporous backing of the Loeb–Sourirajan-type asymmetric membranes which have been used for some commercial separations of gases. He presented a simplified theoretical model of transport on the basis of (activated) Fickian diffusion through the top skin as well as the transition region and Knudsen diffusion only through the microporous backing. This description has been considered to be too simplistic not only for pore flow but also from the point of view of diffusive flow in that "even in the simplest models of membrane transport, Fick's law must be combined with an assumption concerning sorption equilibria at the membrane surfaces, and Henry's law is commonly used for this purpose. But neither Fick's nor Henry's law are adequate in many polymer–permeant systems. . . ." (20). Sirkar derived theoretical expressions to predict membrane performance which were substantiated by data obtained by Gantzel and Merten (21) for pure gases. In another paper, Sirkar (22) described a theoretical optimization of the dense skin thickness for the maximum extent of separation.

The types of membranes described above have been manufactured in spiral-wound module form (23) and used to separate "acid gases" from "sour" natural gas streams containing carbon dioxide which can be used in adjacent oil fields for enhanced oil recovery (24, 25). These membranes were initially developed for the separation of hydrogen from coal gasification processes, but applications for the separation of carbon dioxide and methane from biogas in landfill operations (aerobic fermentation of biomass), separation of oxygen and nitrogen (enrichment of air), dehydration of gas streams, as well as purification and recovery of hydrogen in a variety of chemical and refinery operations have been discussed or demonstrated. It is important to note that while solution–diffusion has been widely agreed to govern the transport and separation of gases through nonporous membranes (24), other workers have described them as consisting of a highly consolidated but very thin microporous skin supported by a relatively thick, highly porous substructure (25). The nature of this thin microporous skin was not discussed but it was considered to provide unusually high permeability while displaying the ability to fractionate molecules of various sizes.

In recent years, the greatest success in membrane gas separations can be attributed to the Monsanto PRISM system, first announced and marketed in late 1979. This is a hollow-fiber system with some physical similarity to the Du Pont Permasep Reverse Osmosis system reported earlier

(26), both having the fibers arranged in a bundle within a vertically oriented tubular pressure vessel which resembles a shell-and-tube heat exchanger. The fibers used in the PRISM separators belong to the class of composite membranes which basically consist of two physically distinct regions, viz., a porous asymmetric substrate region and a coating region. The substrate fibers are generally spun from a so-called "permselective" material such as polysulfone and have a relatively dense skin on the exterior surface which is coated by the addition of a thin, nonselective and highly permeable film (27–29). This discovery earned Monsanto the 1981 Kirkpatrick Chemical Engineering Achievement Award (30). The PRISM separators have found large-scale commercial application in hydrogen purification and recovery in the petrochemical industry and in ammonia production (31–33). Other applications include carbon dioxide recovery in enhanced oil recovery operations and in the upgrading of methane (34, 35).

The hollow-fiber composites offer several advantages (28) compared with the conventional ultrathin coating composites of the silicone/polycarbonate type formed by spreading solutions of the copolymer on water surfaces, which are readily applied to microporous support materials, as reported by Ward et al. (36). The latter and similar membranes have been incorporated into small plate-and-frame modules as portable oxygen-enriching units for use by people suffering from respiratory problems (37). Another technique for preparing composite membranes of cellulose triacetate/polyamide has been described by Kuehne and Friedlander (38) who studied the separation of sulfur dioxide from nitrogen.

RECENT DEVELOPMENTS AND MECHANISMS IN MEMBRANE GAS SEPARATIONS

In the PRISM separators, the coating or the outer film is believed to fill the pores left in the substrate fiber, thus eliminating "leaks." And, since the coating is thin and made from a highly permeable material such as silicone rubber or polyethylene, the permeability of the composite fiber membrane is only slightly less than that of the substrate fiber, while the separation factors exhibited are close to those inherent to the substrate membrane itself having a high "permselectivity." The behavior of these composite hollow fiber membranes in which "the substrate (not the coating) becomes the effective separating material after the coating has been applied" was explained by Henis and Tripodi (28) in terms of their "semi-empirical" Resistance Model (RM) approach. They described the porous asymmetric substrate as consisting of three distinct regions: the surface region or the skin having a certain thickness, the defects or pores in this

skin region, and the highly porous matrix region. Each of these regions, as well as the coating itself, offers resistance to the flow of gases across the membrane in a manner "analogous to resistances in electrical flow."

Henis and Tripodi (28) demonstrated how one would use the model to describe the performance of an RM composite membrane under different conditions, viz., of coating thickness, surface porosity, depth of penetration of coating (separation layer), resistance due to the porous matrix, etc. They noted that "a critical factor in the behavior of RM composite membranes is the fractional area (surface porosity) represented by the pores at the surface, and that the resistance of the pores has been substantially increased by the coating material which has a lower permeability" (compared to the pore itself). As a result, a greater proportion of the gas flow is through the polymer matrix; hence the separation factor of the multicomponent membrane is much closer to the determined intrinsic separation factor of the substrate. This should not, however, imply that an increase in the relative resistance to flow through the pores in turn "forces" more of the permeate gas to flow through the membrane material, as noted to explain the behavior of some posttreated membranes which were believed to have reduced pore cross-sectional area (29). This explanation does not appear to be valid because of the fact that the coating is usually more permeable than the substrate, which means that the resistance of the coating material is less than that of the substrate material; therefore, a greater proportion of the gas flow through the substrate is merely a result of the decreased flow through the pores and not of forcing any additional gas through the substrate material in the skin region. Indeed, the substrate is the separating material owing to its inherent interaction properties with the gas(es), but this is undermined in a highly porous membrane.

Henis and Tripodi (28) defined separation factor as the simple ratio of the individual permeabilities, as has become customary (a common practice) in expressing the selectivity of the so-called homogeneous membranes. They treated the permeabilities essentially as constants arising out of an expression for the permeation rate or flux, similar to a hydrodynamic permeability coefficient in Darcy's law which is not free from the limitation of showing pressure dependence, especially for the transport of compressible fluids through small pores (39). According to the RM approach, the permeation rate and the permselectivity of a composite membrane can be predicted from the intrinsic permeabilities and physical properties of the substrate membrane and the coating materials. Comparison between the experimental separation factors and the predicted ones have not been reported, and while there are agreements between the separation factors, as defined, there are also considerable differences (29). It may be noted

that in the case of homogeneous polymeric membranes, the permeability coefficient, which is essentially a mass transfer coefficient for transport across a membrane, is generally regarded as the product of solubility and diffusivity of the particular gas concerned, the latter being characteristic of transport inside the polymer matrix only. However, Stern and Saxena (40) described the concentration-dependent transport of gases and vapors in glassy polymers, and concluded that the separation of a multicomponent mixture depends on the composition of the mixture, even under the assumption that the dissolved penetrants do not interact with each other, and that maximum separation can be achieved by adjusting the total pressure.

GAS TRANSPORT AND SEPARATIONS THROUGH POROUS MEMBRANES

For all the mechanisms of gas transport through porous membranes, which include Knudsen, slip, viscous, and surface flow mechanisms, it has been demonstrated that the surface flow mechanism contributes significantly to the separation of a gas mixture (41). Also, it is now well recognized that the performance of an asymmetric membrane formed by the phase-inversion technique, followed by appropriate drying or coating procedures for the separation of gaseous mixtures, is strongly affected by variables involved in the membrane preparation procedure. There are, however, only a limited number of reports in the literature in which the effect of membrane preparation variables on membrane performance for gas separation has been systematically studied (42, and references cited therein). In the formation of cellulose acetate membranes, it was found that evaporation time, shrinkage temperature, and solvent(s) used for the replacement of water in the wet membrane during the drying process are some of the important factors which affect the ultimate pore size and pore size distribution of the dry membrane (43). The effect of shrinkage temperature and solvents used in the preparation of the dry membrane on the separation of hydrogen–methane and carbon dioxide–methane gas mixtures with cellulose acetate membranes was further investigated (44–46). A similar study was made by Ohya et al. (47) with respect to the permeation of pure gases and the separation of several gas mixtures through cellulose acetate membranes.

Inagaki and Ohkubo (48) reported a plasma polymerization of hexafluoropropane/methane mixtures and composite membranes for gas separations. More recently, Chen et al. (42) studied the preparation and gas permeation properties of silicone-coated dry polyethersulfone composite membranes. The effect of pore size of the porous polyethersulfone

sublayer and the thickness of the silicone coating on the performance of the coated membranes was examined. Data analysis was applied to the Resistance Model approach of Henis and Tripodi (28), and it was found to be compatible with the conclusions drawn qualitatively from the gas permeation experimental data. It was concluded that a thicker silicone coating is necessary to cover the pores on the surface of the polyethersulfone sublayer as the pore size increases, whereas a considerable amount of gas permeates through incompletely closed pores.

In describing the membranes considered porous to gases and which cannot effect separation, Henis and Tripodi (28) indicated that "such pores need only be 5–10 Å in diameter to permit the passage of most permanent gases across the membrane without actual permeation through the polymer." The existence of such pores was not otherwise substantiated, but this may in fact originate from improper coating or the coating itself, especially when the coating material is also of high molecular weight and may not be in "occluding contact" with a polymeric substrate membrane which is highly porous. It is notable that Henis and Tripodi (29) illustrated several examples of multicomponent membranes which exhibited various separation factors, sometimes lower than the determined intrinsic separation factor of the coating material, which resulted from imperfections in the coating itself.

In the case of highly shrunk porous cellulose acetate membranes, Glueckauf (49) reported that the pore diameters are of the order of 10 Å, while Agrawal and Sourirajan (14) estimated, on the basis of Liepmann's analysis (50), average pore diameters varying from 6.4 to 38 Å depending on the gas permeating through such membranes. Methods based on gas permeation through microporous polymeric membranes have also been described by other workers (51–54), but these have dealt with membranes with relatively large pores, and the applicability of the methods has been criticized (55). However, regardless of the magnitude of the pore size which may be suitable for effecting any gas separation, there is no quantitative model which can predict the performance of a membrane in the permeation and separation of gases on the basis of all the various transport mechanisms.

We have previously reported (56) a quantitative model which takes into account all the gas transport mechanisms through porous media and allows one to calculate the average pore size and pore size distribution of the membrane based on experimental permeation data for a reference gas. An attempt was made to predict the total gas permeation rate and separation factor on the basis of transport equations developed in this work, and it was found that the average pore diameters of dry cellulose acetate

membranes which allowed reasonably good separation of hydrogen–methane gas mixtures are typically in the range of 6 to 14 Å (41). The importance of pore size and the different mechanisms involved in the permeation and separation of gases through dry cellulose acetate membranes was also demonstrated clearly, albeit qualitatively, by Lui et al. (46). In studying the solvent exchange technique for making dry cellulose acetate membranes, these workers concluded that there is a critical pore size on the surface of wet membrane that results in the smallest pore size on the dry membrane and consequently in the highest separation factor of the membrane used for gas separation.

It can be noted that the pure gas permeabilities of asymmetric porous cellulose acetate membranes reported in our earlier work (41) are comparable to those reported in the literature for other gas separation membranes. For example, the experimental permeability coefficient of pure methane gas using the membrane designated CA-33 was 0.22×10^{-12} kmol/($m^2 \cdot s \cdot Pa$) which, in terms of conventional units, is equivalent to 0.66×10^{-6} cm^3(NTP)/($cm^2 \cdot s \cdot cmHg$). This compares very well with that of 0.7×10^{-6} cm^3(NTP)/($cm^2 \cdot s \cdot cmHg$) obtained for the so-called asymmetric dense cellulose acetate membrane, which is several orders of magnitude higher than the value of 0.14×10^{-10} cm^3(NTP)/($cm^2 \cdot s \cdot cmHg$) reported for a completely dense and homogeneous membrane by Gantzel and Merten (21). Similarly, a multicomponent membrane consisting of porous cellulose acetate coated with poly(dimethylsiloxane) has been reported by Henis and Tripodi (29) to have a hydrogen permeability of 1.5×10^{-5} cm^3(NTP)/($cm^2 \cdot s \cdot cmHg$) using a mixture of hydrogen and carbon monoxide. This is again identical to our result for pure hydrogen gas permeability of 4.64×10^{-12} kmol/($m^2 \cdot s \cdot Pa$) or 1.4×10^{-5} cm^3(NTP)/($cm^2 \cdot s \cdot cmHg$) obtained for the CA-33 membrane which had an average pore diameter of 10 Å.

Another important feature of the dry gas separation membranes used in our studies is their surface porosity which can be calculated on the basis of results from gas transport equations described earlier (56). Using an average pore diameter of 10 Å and assuming an asymmetric layer thickness of 1000 Å, the latter being considered typical of asymmetric hollow fiber membranes by Henis and Tripodi (28), the total number of pores, and hence the total pore cross-sectional area for the CA-33 membrane, could be calculated to be 3.4×10^{-9} m^2. According to the terminology and definition of Henis and Tripodi (28), the surface porosity is obtained by dividing the total pore cross-sectional area by the effective membrane area, which gives 0.33×10^{-5} or $33 \times 10^{-5}\%$. This is at least two orders of magnitude higher than the surface porosity of $10^{-7}\%$ which is considered minimum for an effective nonporous membrane (28).

CONCLUSIONS

It appears that an ideal membrane for gas separation should contain a large number of sufficiently small but asymmetric pores in order to yield a reasonably high flux as well as a good selectivity (separation factor). The interplay of different mechanisms of transport through such porous membranes leads to various trends in permeabilities as a function of pressure (56). This is also expected to apply in the case of a separation factor which generally increases with increasing pressure owing to the preponderance of surface flow over pore flow (14). However, because of limitations in the extent of adsorption on the pore surface, depending on gas–membrane interaction and the available pore size for gas-phase transport, as well as on the mobility of the adsorbed layer, it may not always be possible to predict the separation factor of a mixture of gases.

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