Catalysis with Inorganic Membranes

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Introduction and Background

Catalytic inorganic membranes are among the most challenging and intriguing porous materials. Consisting of a thin film of mesoporous or microporous inorganic material deposited on a macroporous material, catalytic membranes are multifunctional materials that must be engineered for both chemical and physical properties. New approaches to carrying out chemical reactions are possible by tailoring the membrane catalytic activity and selectivity, permselectivity, and other thin film properties. Readers are referred to several recent reviews of inorganic membranes, in particular, Zaspalis and Burggraaf,[1] Armor,[2] Gellings and Bouwmeister,[3] Hsieh,[4] Stoukides,[5] and Tsotsis et al.[6]

Inorganic membranes are most conveniently classified according to pore size (see introductory article). Of particular importance is the ratio of the pore size to the molecular mean free path (MFP). Decreasing pore dimensions lead to increased selectivity with corresponding loss of permeability. *Macroporous* membranes have a pore size much larger than the MFP, leading to molecular (bulk) diffusion or viscous flow. Knudsen diffusion dominates in the *mesoporous* regime, where the pore size is comparable to the MFP. In addition, surface diffusion of the molecules along the pore walls may contribute, leading to an enhanced flux of the adsorbed species along the walls. The *microporous* regime is encountered when the pore size is comparable to the molecules. This regime makes possible much higher permselectivities, which depend on both molecular size and specific interactions with the solid. Finally, in dense membranes, molecular transport occurs through a solution-diffusion mechanism, which also involves specific interactions between the solute and membrane.

Catalytic inorganic membrane reactors are classified according to the type of membrane (permselective versus nonpermselective) and the location of the catalyst (within the membrane versus external to the membrane). Figure 1 shows four common catalytic membrane types, following the notation of Tsotsis et al.[6] The catalytic membrane reactor (CMR) consists of a support layer and a permse-

Permselective Active Layer
Nonpermselective Active Layer
Permselective Layer without Activity
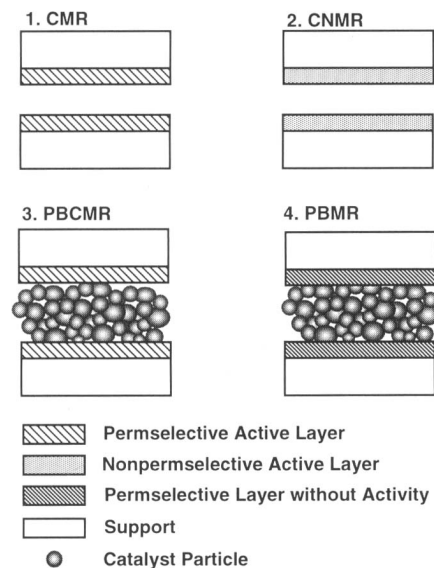Support
O Catalyst Particle

Figure 1. Four different membrane reactor types: catalytic membrane reactor (CMR), catalytic nonpermselective membrane reactor (CNMR), packed bed catalytic membrane reactor (PBCMR), and packed bed membrane reactor (PBMR).

lective layer (membrane) with a catalytic function. The catalytic nonpermselective membrane reactor (CNMR) consists of a support layer and a nonpermselective catalytic layer. In the packed-bed membrane reactor (PBMR) the catalyst is located external to the supported permselective membrane. In the packed bed catalytic membrane reactor (PBCMR) the catalyst is located external to the permselective membrane which itself has catalytic activity.

This review describes the working concepts of several successful applications of catalytic inorganic membranes. We identify the materials requirements of the membrane, discuss how the combination of membrane permeation and catalysis can result in a device with capabilities not achievable with more conventional chemical reactor designs, and describe how certain membrane characteristics affect the performance of the reactor.

Reactions Requiring Strict Stoichiometric Feeds

A class of catalytic reactions requires precise control over the feed composition. Consider the general reaction given by

$$A + \nu_B B \rightarrow \nu_P P + \nu_Q Q. \tag{1}$$
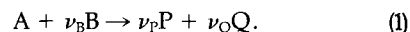
A notable example is the Claus reaction

$$SO_2 + 2 H_2S \leftrightarrow 3/8 S_8 + 2 H_2O. \tag{2}$$
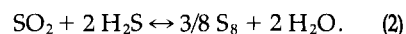
One could feed a conventional reactor with a stoichiometric ratio of reactants A and B, i.e., $1/\nu_B$. If there is a change in the supply of either reactant, however, then "slip" of the excess reactant to the reactor effluent stream will occur. Slip of H_2S must be avoided in the example reaction because it is a toxic pollutant.

A nonpermselective catalytic membrane can effectively minimize reactant slip.[7,8] The idea is to flow, for the general reaction above, reactants A and B on opposite sides of a porous catalytic membrane (Figure 2). The key to the concept is that the feed flow rates of A and B need not be adjusted to match the needs of stoichiometry.

Suppose the membrane reactor consists of a porous tube separating the flowing A and B streams. Moreover, suppose the reaction between A and B is instantaneous and irreversible. At a given position along the length of the membrane reactor, A and B are consumed completely at an interface within the membrane. In a nonstoichiometric, steady-state situation, the reaction interface will be located so that the local concentrations of A and B vanish and the supply fluxes of A and B match the stoichiometric requirements. Thus, slip of either A or B to the opposite side of the

membrane is prevented. One can envision a situation in which one of the reactants (say, reactant B) could be continuously recirculated on one side of the tubular membrane in such a way that complete conversion of reactant A is achieved with negligible slip of reactant B. An added feature is the prevention of possible side reactions between, for example, species B and a component in the A stream.

The concept has been demonstrated experimentally.[7-9] The results from these pioneering studies suggest that the following materials properties be satisfied to successfully prevent reactant slip:

(1) The membrane may consist of a single catalytically active layer or of a support layer and catalytic layer;

(2) The catalytic layer must have a sufficiently high activity; and

(3) The membrane must have a sufficiently low overall permeability but does not require a permselective function.

The combination of 1, 2, and 3 guarantees that the membrane is operated in the diffusion-controlled regime. The membrane thickness must be tailored according to the reaction kinetics.

Rate Enhancement of Volatile-Reactant-Limited Multiphase Reactions

Nonpermselective inorganic membranes can be used in multiphase catalysis. The general reaction is of the form

$$A_{(G, L)} + B_{(L)} \rightarrow P_{(L)} \qquad (3)$$
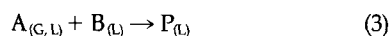
where G (L) denotes gas (liquid). Species A represents the more volatile reactant, which may be only sparingly soluble in the liquid phase consisting of less volatile reactant B, and product P. An example reaction is the hydrogenation of an unsaturated hydrocarbon on a supported noble metal catalyst.

In a conventional trickle-bed reactor, gas and liquid flow concurrently downward through a bed of catalytic pellets. Catalysis occurs on the liquid-covered active sites. The supply of sparingly soluble reactant A to the sites is inhibited by the liquid containing B and P,[10] leading to under-utilization of the catalyst. The hollow tube, multiphase, inorganic membrane reactor[11] is a more efficient alternative.

A schematic of the concept is provided in Figure 3. A hollow, macroporous ceramic tube is coated on its inside wall with a thin, catalyst-impregnated mesoporous ceramic membrane. The gas and liquid phases are segregated on opposite sides of the catalytic membrane. Because of capillarity, the liquid penetrates the support and membrane, creating a gas-liquid inter-
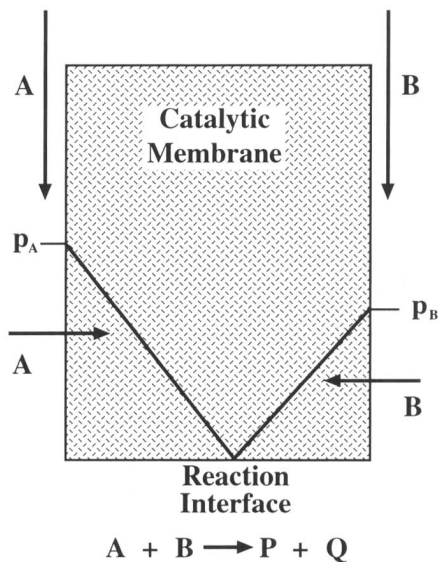

Figure 2. Nonpermselective membrane system for rapid gas-phase catalytic reactions requiring strict stoichiometric feeds.
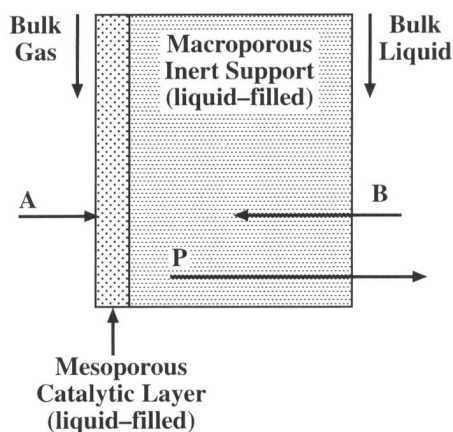

Figure 3. Membrane system for rate enhancement of a multiphase catalytic reaction.

face near the inside wall. Both the liquid and volatile reactant flowing through the tube core are in direct contact with the catalyst. The key is that the volatile reactant does not have to diffuse through a liquid film, as in the trickle-bed reactor.

The concept was demonstrated by Cini and Harold[11] for the hydrogenation of α-methylstyrene to cumene on a Pd-impregnated γ-Al$_2$O$_3$/α-Al$_2$O$_3$ membrane. For a hydrogen presaturated liquid feed, the overall rate (normalized by the

amount of active material present) for the tubular membrane was 5–7 times that for the fully wetted single pellet. The overall rate with the tube was 15–20 times higher than for the pellet exposed to an unsaturated liquid.

The inorganic membrane in this application has the following materials attributes:

(1) Membrane thickness up to several hundreds of microns,

(2) Nominal pore size in the mesopore range (20–500 Å),

(3) Specific surface area up to 200 m^2/g,

(4) Capability to precisely position the catalytic component within the film and with a high loading, and

(5) Good adhesion of the film to the support.

(6) Defects and pinholes can be tolerated as long as (4) is satisfied.

The exact values of film thickness, pore size, surface area, and catalyst loading and composition depend on the reaction. For example, the film thickness depends on the catalytic activity; the goal is to have as thick a film as possible without encountering severe diffusional limitations.[12]

Equilibrium-Limited Reactions

Another application area for membrane reactors involves reversible reactions given by

$$aA \leftrightarrow bB + cC. \qquad (4)$$

An important example is catalytic dehydrogenation, where A, B, and C would represent the hydrocarbon, dehydrogenated hydrocarbon, and hydrogen, respectively.

Membranes that are permselective to one of the products offer a way to confront the constraint imposed by the reaction equilibrium. The idea is to selectively remove product, thereby shifting the equilibrium. The successful performance of this class of membrane reactor relies on a membrane that has the following basic materials properties:

(1) The membrane should have a sufficiently high permselectivity to the targeted reaction product;

(2) The catalytic activity of the membrane itself or of the catalyst external to the membrane must be sufficiently high;

(3) The membrane must withstand moderate to high temperatures (400–700°C);

(4) If the membrane is completely permselective to the targeted reaction product, the overall permeability should be as high as possible; and

(5) If the membrane is not completely permselective to the targeted reaction

product, the overall permeability should be confined to an intermediate range.

The basis for property (1) is that selective product removal facilitates the shifting of equilibrium. The operating principle requires that the reaction be operated close to equilibrium, thus the need for properties (2) and (3). Justifications for properties (4) and (5) follow.

For hydrogen-producing reactions in particular, two types of hydrogen-permselective membranes, dense and porous, have been investigated. The dense membranes are typically metallic, usually containing Pd or Pt.[13] The metallic membranes can be in the form of hollow tubes or foils, or films supported on a porous substrate prepared by various methods.[14-16] Porous membranes include both the mesoporous Knudsen variety, principally γ–Al_2O_3 and Vycor® glass. Thin-film ceramic membranes can be prepared by sol-

Figure 4. Simulation of cyclohexane dehydrogenation: The conversion of cyclohexane is plotted as a function of H, the ratio of the permeation rate of hydrogen to the characteristic reaction rate.[27] Curves 1 and 2 correspond to a mesoporous membrane with Knudsen separation factors. Curves 3 and 4 correspond to a microporous membrane with a separation factor of 100. Curves 5 and 6 correspond to a microporous membrane without defects with an infinite separation factor. Odd (even) numbered curves indicate an inert feed rate on the permeate side that is equal to (10 times that of) the cyclohexane feed rate on the tube side. For comparison, a University of Twente-designed Knudsen membrane is indicated by the lower right rectangle. Two microporous SiO_2/Al_2O_3 membranes are shown by the other rectangles.

gel coating techniques[12,17-24] or chemical vapor deposition (CVD).[25,26] Generally, Knudsen membranes offer reliable performance and high permeability but limited permselectivity. Dense membranes offer high permselectivity, limited permeability, and unproven reliability, the last a result of coking and embrittlement.

Conversion efficiency depends critically on the relative hydrogen permeation rates through the membrane.[27] Figure 4 shows model calculations for a tube-type PBMR system for dehydrogenation of cyclohexane to benzene using hydrogen-permeable membranes of differing selectivity, including a mesoporous Knudsen membrane (curves 1, 2), a selective microporous membrane with some defects (curves 3, 4), and an infinitely selective microporous membrane (curves 5, 6).

Several trends in Figure 4 deserve discussion. The cyclohexane conversion is nearly independent of selectivity at low permeation rates even though overall conversion exceeds the equilibrium value of 8% (based on the tube-side feed). The membranes with finite separation factors (curves 1–4) exhibit a maximum in the conversion. The decrease at high permeation rates is due to loss of reactant (cyclohexane) from the reaction zone to the permeate side.

Using the principles captured in Figure 4, ceramic membranes of both SiO_2 and SiO_2/TiO_2 have been synthesized at the University of Twente.[19,23,24] Selected membranes are shown in Figure 4. The synthesis involves dip-coating of polymeric sols, followed by drying and calcination steps. The dip-coating procedure yields films with thicknesses in the range of 300–1000 Å. Permeability analyses of the SiO_2 and SiO_2/TiO_2 membranes clearly indicate an activated transport regime that is characteristic of microporosity.[23,24] For the SiO_2 membrane with defects (selectivity factor of 100) nearly optimal performance is achieved. For the SiO_2 membrane without defects (infinite selectivity factor; curves 5 and 6) the performance is excellent. The combination of reasonable permeability and high permselectivity makes the sol-gel derived SiO_2 an attractive candidate for equilibrium-shift applications.

Methods other than the sol-gel coating are being developed for synthesizing permselective ceramic membranes for the equilibrium-shift application. Gavalas et al.[25] developed a CVD technique for supported microporous oxides. An oxide layer is deposited at a precise location within a porous support. More recently, the hydrolysis of chlorides in porous glass has been used to produce permselective SiO_2, Al_2O_3, and TiO_2 layers.[28] Although the

permselectivity is excellent, the permeability is well below that of the sol-gel derived SiO_2 membranes just described. Finally, Ma, Dixon, and co-workers[29] developed hollow glass fiber materials. They achieved a H_2/N_2 separation factor of 84 with permeabilities comparable to sol-gel derived membranes.

Dense metallic membranes require as high a permeability as possible. Based on Figure 4, Pd films less than 1 μm are required to be competitive with the microporous films just described. An alternative is to increase the reactor length or reduce the reactant flow rate to achieve higher conversions. This strategy, however, reduces the volume efficiency of the reactor.

Successful utilization of the equilibrium shift by selective permeation requires that the catalysis be sufficiently fast that reaction equilibrium is approached. Raich and Foley's[30,31] simulations of a Pd membrane reactor underscore this point. Literature kinetics data for cyclohexane dehydrogenation on 0.5% Pt/Al_2O_3 catalyst and isobutane dehydrogenation on 20% Cr_2O_3/Al_2O_3 were used for a comparison of both reactions. The results showed that an equilibrium shift occurred with the cyclohexane system but not with the isobutane system. The key difference between the two is the relative rates of reaction. The cyclohexane dehydrogenation rate on the Pt catalyst is sufficient to approach equilibrium. Thus the selective permeation of hydrogen can be exploited. The isobutane dehydrogenation is kinetically limited so the equilibrium shift cannot be applied.

Recent developments in the synthesis of metallic membranes include the development of Pd-based alloys and supported metallic membranes. Notwithstanding the high permselectivity of Pd to hydrogen, Pd membranes are known to suffer from embrittlement due to repeated hydrogen sorption and desorption cycling.[13] Certain Pd-based alloys show improved durability and have a high hydrogen permeability.[19,20,32] Edlund and co-workers[33,34] developed a multilayer metallic membrane. The device has several novel features. It consists of a metallic support layer and a thin permselective metallic layer separated by an oxide to prevent interdiffusion of the metals. This flexible structure can withstand temperatures up to 700°C where substantial permativity enhancement is realized because of the activated nature of hydrogen transport.

Finally, a note of caution is advised in assessing the performance of a membrane reactor in the equilibrium-shift application. Ideally, one should treat the membrane reactor as an isolated system. The single input to the system is the combina-
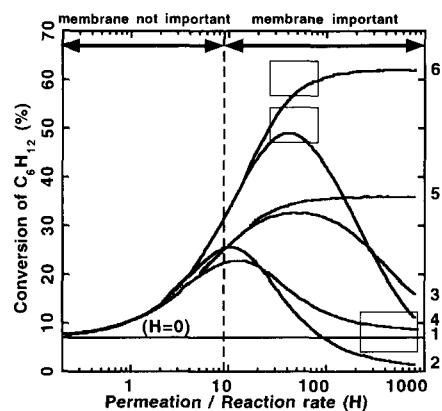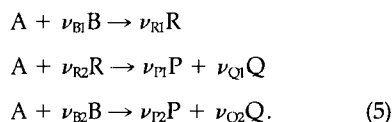
tion of the tube-side and shell-side feeds. One should then compare the performance of a conventional catalytic reactor operated with such a feed to that of the membrane reactor. For example, dilution of a feed to a conventional fixed-bed reactor can increase the equilibrium conversion. The fixed bed operated with a diluted feed should then serve as the benchmark for comparing the membrane reactor. This insures an unambiguous assignment of any conversion increase that is attributed to the selective permeation concept.

Consecutive-Parallel Reaction Systems

One of the continuing challenges in reactor engineering is to maximize the yield of a desired component in a multiple-reaction network of the form

$$A + \nu_{B1}B \rightarrow \nu_{R1}R$$

$$A + \nu_{R2}R \rightarrow \nu_{P1}P + \nu_{Q1}Q$$

$$A + \nu_{B2}B \rightarrow \nu_{P2}P + \nu_{Q2}Q. \qquad (5)$$

In many partial hydrogenations and oxidations, R would represent the desired product and P the undesired product. An example is the reaction between oxygen (A) and ethylene (B) to produce desired acetaldehyde (R) and undesired carbon oxides (P) and water (Q).

In consecutive-parallel partial oxidation reactions, membranes are used to limit the oxygen (A) supply.[3,5,35,36] Feeding the oxidant, A, from the support side achieves a low effective A/B ratio in the reaction zone. This approach can result in higher desired product selectivity if certain kinetic ingredients are satisfied.[36]

Dense Membranes for Partial Oxidation

The segregated feed condition can be achieved using a solid electrolyte such as Y_2O_3-stabilized ZrO_2 (YSZ) or perovskites.[37-39] These materials have virtually infinite permselectivity to oxygen.

The concept of the dense oxide membrane reactor for partial oxidation is as follows. Air is flowed on one side of a membrane device, which consists of a macroporous support and a dense oxide layer that can selectively permeate oxygen. Hydrocarbon is flowed on the other side. The permeation of oxygen through the oxide lattice provides a controlled supply of lattice oxygen to the other side where the catalyst and flowing hydrocarbon are located. The catalyst is either the permselective oxide layer itself or is another material that is deposited in particulate form on top of the membrane layer.

Besides providing a controlled supply of oxygen, an additional attractive feature is that the lattice oxygen may be a more selective form—in a kinetic sense—for hydrocarbon partial oxidation reactions.[3,40-42] Gas phase oxygen and adsorbed oxygen are known to react with the hydrocarbon to form undesirable carbon oxides in addition to the partly oxidized hydrocarbon. Thus, if the supply of lattice oxygen can be properly balanced with its rate of consumption to the desired species, this approach enables a sustained oxidation of the hydrocarbon without mixing the hydrocarbon and air.

Several experimental studies have been carried out with success. In particular, methane conversion to higher value products (e.g., higher hydrocarbons, synthesis gas) has been a keen subject of interest. An advantage of the methane partial oxidation reaction system is that the high temperature needed to get reasonable oxygen flux is compatible with the temperature needed to activate the methane for reaction. Omata et al.[43] demonstrated with a PbO/MgO membrane an open-circuit selectivity of 97% to C2 hydrocarbons. Hazbun[40] reported 10–50% methane conversion and higher hydrocarbon selectivity exceeding 50% at 850°C. The inside wall of a porous hollow tube was coated with a TiO_2-doped YSZ layer. A mixed metal oxide layer served as the catalyst. Other investigators have employed electrochemical pumping of oxygen.[35,44]

Insight into the performance of an oxygen-permeable dense membrane reactor for methane oxyreforming was provided by Tsai et al.[45] The model included the combustion and reforming reactions given by

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$

$$CH_4 + H_2O \rightarrow CO + 3H_2$$

$$CH_4 + CO_2 \rightarrow 2CO + 2H_2. \qquad (6)$$
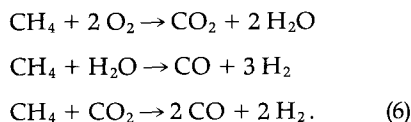
The overall reaction, the partial oxidation of methane to synthesis gas ($CO + 2H_2$) is exothermic. The model comprised a plug-flow type reactor consisting of a shell side where air is fed, a catalyst-packed tube side where the methane is fed, a porous catalytic layer, an oxygen-permeable dense layer, and an inert support layer. Literature kinetics and permeability data were used to handle the reaction rates and the oxygen transport through a perovskite ($SrCo_{0.8}Fe_{0.2}O_{3-x}$). Heat effects were accounted for in the model. Selected simulations demonstrate the effectiveness of the membrane reactor in controlling the reactor temperature. This control is a con-

sequence of a reduced rate of heat generation per unit reactor volume due to the oxygen transport limitation.

A revealing study was carried out by Di Cosimo et al.[42] for propylene oxidative dehydrogenation to the dimers 1,5-hexadiene and benzene. Nonselective products for this reaction are methane, carbon oxides, and water. The ion-conducting dense membrane, $(Bi_2O_3)_{0.85}(La_2O_3)_{0.15}$, served as the catalyst in this example. The investigators compared the performance of the segregated flow case, in which oxygen was continuously supplied through the 38 μm thick membrane, to mixed flow cases. For example, the segregated flow case exhibited the highest desired product yield (0.024). When a propylene-rich feed (20% propylene, 1% O_2) was fed continuously to a fixed bed of the same material, the yield was about 40% lower. The investigators concluded that the lattice oxygen is that which is involved in the selective oxidation.

Despite these demonstrated successes, selective membranes for partial oxidation face hurdles. The largest hurdle is the limited flux of oxygen at temperatures compatible with most partial oxidation reactions (<600°C). There is an incentive to synthesize thinner oxygen-conducting membranes.

The second hurdle is reliability. Cracks must be avoided. This hurdle is a challenging one, especially if thinner membranes are sought for the higher fluxes.

The third hurdle is to correctly match the membrane with the catalyst. That is, to exploit the membrane concept, the rate of oxygen supply should be limiting. Otherwise, oxygen could recombine on the catalyst side and form the potentially less selective molecular form. On the other hand, too low an oxygen supply is bound to have detrimental economic consequences.

Strategies that remove the desired product R would also be effective, following the product removal strategy of the previously mentioned equilibrium-shift concept. This strategy would require, for example, a dense or microporous membrane more permeable to a partially oxidized hydrocarbon than to both the unoxidized hydrocarbon and to oxygen, a formidable materials challenge.[46,47]

Nonpermselective Membranes for Partial Oxidation Reactions

This section describes selected experimental and modeling results that demonstrate the concept of the nonpermselective membrane reactor with reactant segregation. Compared to the dense membrane approach, this approach offers higher oxygen flux, but limited permselectivity.

Harold et al.[48] used the partial oxida-

tion of ethylene to acetaldehyde on an alumina-supported vanadium oxide membrane. The membranes used consisted of 4 nm mesoporous γ-Al_2O_3 films supported on top of macroporous α-Al_2O_3 substrates. The overall experimental system was similar to that used for previous studies.[49-51]

Membrane reactor tests consisted of two flow types. In the *mixed feed* experiments a mixture of ethylene, oxygen, and helium was fed to the active layer side of the membrane reactor while pure helium was fed to the support side. In the *segregated feed* experiments, a mixture of ethylene and helium was fed to the active layer side while a mixture of oxygen and helium was fed to the support side. Figures 5 and 6 show the results of a set of experiments in which the flow rates of ethylene and oxygen were fixed (equimolar feed) and the reactor temperature was varied.

The mixed flow results were found to be similar to those obtained for a conventional packed-tube of catalyst. At low temperature the major products are CO and acetaldehyde. At high temperatures the major product is CO_2. As shown in Figure 5, the maximum acetaldehyde selectivity observed was 20% at an intermediate temperature of 220°C. The segregated feed experiments indicate a qualitative but not quantitative similarity to the mixed feed results (Figure 6). The maximum selectivity to acetaldehyde is approximately 65%, considerably higher than for the mixed flow. This maximum occurs at a higher temperature (around 310°C) than that obtained for the mixed feed.

The observed higher acetaldehyde selectivity for segregated feed demonstrates the principle of reactant segregation. Controlling the flux of oxygen to the active layer maintains a higher ethylene to oxygen ratio. In effect, a diffusion limitation (for oxygen) is created. This limitation is beneficial to acetaldehyde selectivity but detrimental to overall rate. While a higher acetaldehyde selectivity was attainable at a fixed ethylene conversion for the segregated feed, a higher temperature was needed to achieve that conversion level. Thus, in sacrificing overall rate for selectivity, a temperature price must be paid to maintain productivity.

Results similar to those obtained with the segregated-feed membrane reactor might be obtained by distributing the feed of oxygen along the length of a conventional fixed-bed reactor.[52] This approach would also maintain a high ethylene to oxygen ratio throughout the reactor. Any reactor comparison study should consider such a reactor design and strategy as an alternative.

Membrane Reactor Design Considerations and Challenges

Catalytic inorganic membrane reactors have potential to replace conventional reactors in certain chemical processes. For applications in which permselectivity is a necessary membrane feature (e.g., equilibrium shift, partial oxidation in dense membrane reactors), it is clear that materials engineering issues are the key. Advances in sol-gel and physical and chemical vapor deposition techniques are essential to confront permselectivity and permeability issues.

For applications in which permselectiv-

ity is not necessary, the membrane reactor design and operating strategy must be fine-tuned to take advantage of the reaction zone defined by the catalytic membrane. Application of reaction engineering principles is necessary here.

Most of the membrane reactors described in this review have an inherent weakness: the overall rate of the desired reaction is diffusion-limited. A diffusion-limited reaction implies that a certain fraction of the reactor volume is not utilized for reaction. This sacrifice of reactor volume for higher desired product selectivity must be economically justified. Because of the diffusion limitation, the area available for transport per unit volume of reactor, the A/V ratio, becomes an important membrane reactor design parameter. Obviously, the higher the A/V, the lower will be the material cost. This can be accomplished by reducing the inner and outer diameter of a tubular support, for example. However, one must consider material strength and pressure drop limitations. Another way is to use more densely-packed architectures. For example, a cellular structure was described by Hazbun.[40]

Most of the reaction systems of interest have large heat effects. The equilibrium-limited reactions are typically endothermic. The partial oxidation reactions are typically exothermic. Thus, efficient means of supplying or removing heat are essential. Most of the research to date has not addressed this critical issue.

High-temperature seals are also important. One must be able to prevent leakage between the two flowing streams. While clever sealing techniques have been demonstrated at the laboratory scale, sealing a commercial-scale membrane reactor remains a challenging engineering problem.

Safety and reliability are crucial intangibles to be faced in the development of membrane reactors. Heat management and sound sealing designs are essential to prevent reactor runaways or the mixing of potentially flammable components. Long-term, reliable performance of a catalytic membrane reactor needs to be demonstrated.

Finally, any proposal to develop a new process based on a membrane reactor deserves close scrutiny. One should consider modifications of conventional reactor designs and operating strategies as well as alternative strategies. In the end, economic factors, reliability, and safety will determine if membrane reactors become a commercial reality.
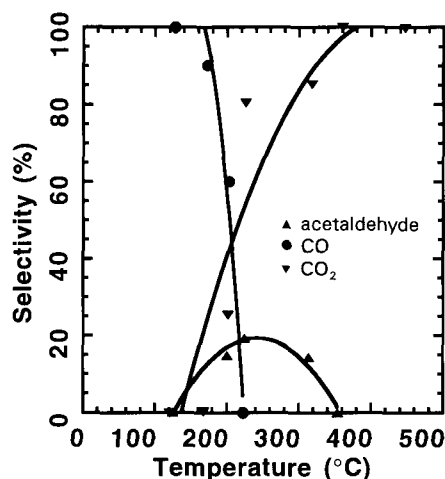
Figure 5. Product distribution obtained with the mixed feed for ethylene oxidation.
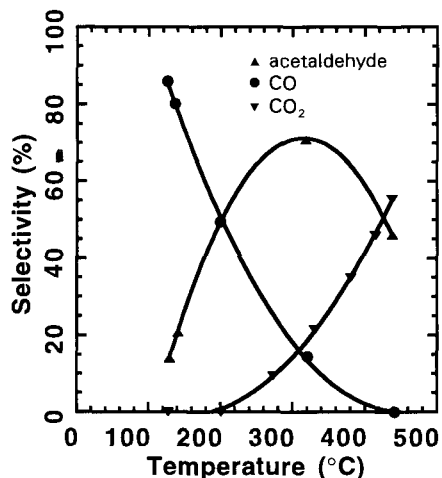

Figure 6. Product distribution obtained with the segregated feed for ethylene oxidation.

membrane group under the direction of Prof. Burggraaf.

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