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Gas/gas membrane contactors – An emerging membrane unit operation

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ABSTRACT

Gas/gas membrane contactors are devices in which a feed gas and a sweep gas are circulated on either side of a membrane. The pressures of the two gas streams are often approximately equivalent, so the principal driving force for permeation is the concentration difference between the feed and sweep gas components. This type of contactor has been commercialized for energy recovery devices in air conditioning applications. More recently, these contactors have been suggested for use in carbon dioxide capture and sequestration. In this paper, the performance of an ideal contactor using perfectly selective membranes is examined. For this type of ideal contactor, analytical equations can be derived that allow the partial pressure profiles within the contactor to be calculated. The effect of the sweep ratio (sweep flow rate/feed flow rate) and feed pressure on separation performance of the contactors has been calculated. In the final section of the paper, the performance of contactors fitted with membranes permeable to other components of the feed gas is described.

1. Introduction

This paper describes the operation of gas/gas membrane contactors. These are typically low-pressure devices in which a feed gas is circulated across one surface of a permeable membrane, and an approximately equal volume of another gas, at a similar pressure, is circulated countercurrently on the other side of the membrane. Permeation occurs because of the partial pressure difference between components in the feed and sweep gases. Most of this partial pressure difference occurs because of concentration rather than pressure differences across the membrane. This type of contactor has been commercialized for energy recovery devices in air conditioning and fuel cell humidity control applications [1–3]. More recently, these contactors have been suggested for use in carbon dioxide capture [4,5].

Most membrane gas separation processes do not use a permeate side sweep gas. In conventional processes, a feed gas mixture flows across the surface of a permselective membrane; a portion of the mixture permeates the membrane and is removed as lower pressure permeate gas. The remaining gas, depleted in the permeating components, is removed from the feed side of the membrane as a residue gas. One stream enters the membrane module (the feed), two streams leave (the residue and permeate).

A few membrane processes have been developed in which a small flow of sweep gas is introduced on the permeate side of the membrane. This gas usually flows countercurrently to the incoming feed. Two streams enter the module (the feed and sweep) and two streams leave (the residue and permeate + sweep). This type of sweep process has been used when the membrane selectivity is much higher than the pressure ratio across the membrane, and a small amount of an extremely permeable component must be removed. A well-known example is the removal of water vapor from compressed air, first commercialized in the 1990s [6,7]. In this process, the membrane performance is pressure ratio limited, and the separation can be much improved by using a small fraction of dry gas as a permeate side sweep to reduce the partial pressure of the permeating component (water) on the permeate side of the membrane. In these applications, the volume of sweep gas used is only a few percent of the volume of the feed gas to be separated.

A few years ago, Membrane Technology and Research, Inc. (MTR) proposed using a gas/gas membrane contactor to separate carbon dioxide (CO_2) from fossil fuel power plant flue gas [4,5]. If successfully developed, this could be an important membrane application. In this process, CO_2-rich flue gas passes across the feed side of a membrane, while an approximately equal volume of air passes as a sweep gas on the membrane permeate side. In this way, the partial pressure of CO_2 on the permeate side is maintained at a lower level than on the feed side. Because of this partial pressure difference, CO_2 passes from the flue gas into the sweep air stream. If membranes are used that are very permeable to CO_2,

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but relatively impermeable to oxygen and nitrogen, very little nitrogen passes from the flue gas into the sweep air and very little oxygen passes from the air sweep into the flue gas. Because the feed and permeate pressures are low, blowers are the only compression equipment needed to circulate the gases across the membrane. A useful separation is performed at a minimal energy cost.

A block diagram illustrating the application of this device to CO_2 capture in a coal-fired power plant is shown in Fig. 1. Air passing countercurrently to the flue gas strips CO_2 out of the gas. The CO_2-laden air is then used to burn coal in the power plant boiler. This combustion process is used to produce steam to drive a turbine and make electricity; it also generates a CO_2-enriched flue gas. A portion of this flue gas is separated as a CO_2-enriched stream by a conventional membrane unit. The remaining gas passes across the feed surface of the membrane contactor and becomes the CO_2-depleted gas (2% CO_2) that is discarded through the chimney. By using the membrane contactor, the CO_2 concentration in the flue gas from the boiler can be enriched from the normal concentration of 10–13% CO_2 to 20% CO_2 or more. Enhancing the CO_2 concentration of the flue gas from the coal combustor increases the efficiency of the selective CO_2 purge step. A more concentrated CO_2 purge is produced because only a portion of the CO_2 must be removed in this step. The remaining CO_2 is recycled by the membrane contactor. The use of a membrane contactor significantly reduces the cost of separating a CO_2 concentrate stream from the flue gas. This process is under development by MTR as a potential CO_2 capture technology.

In this paper, a gas/gas contactor of the type shown in Fig. 1 is treated as a unit operation. The impacts of different process parameters on unit performance are examined. The paper is divided into two sections. In the first section, we will show calculations for an ideal contactor; that is, a contactor fitted with a membrane permeable to one of the components of the feed gas (CO_2), but impermeable to all other components. We will also assume the component to be removed is present at a low concentration ($\sim$1%) so that the volume change in the feed and sweep streams caused by permeation can be ignored. The properties of a gas/gas contactor will then be illustrated using this ideal device.

In the second section of the paper, we will illustrate the complications that result when real membranes with limited selectivity are used and permeation of other components is possible. The changes in performance that occur when permeation through the membrane causes volume changes to the feed and sweep flows will also be examined. This type of contactor is closer to the type that would be used for the application shown in Fig. 1.

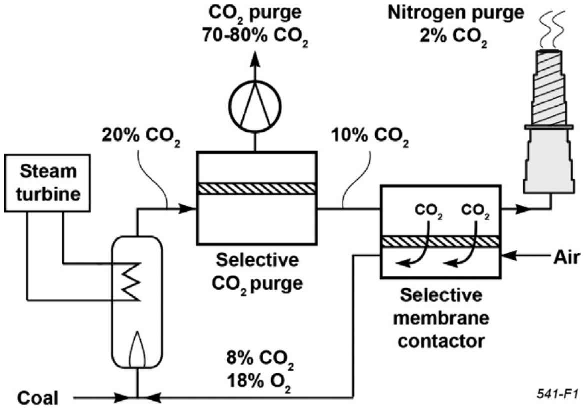

Fig. 1. Block diagram illustrating the use of a selective membrane contactor to recycle CO_2 to the boiler of a coal power plant. In this way, the concentration of CO_2 in the flue gas exiting the boiler increases from 10–13% to 20% at very little energy cost [4].

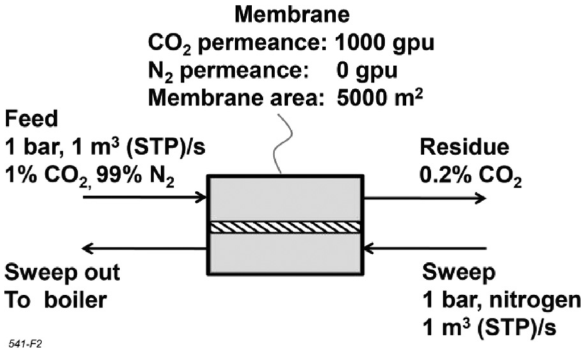

Fig. 2. Base case conditions used in this paper for an ideal gas/gas membrane contactor.

2. Ideal contactor performance

The base case operating conditions for an ideal contactor that forms the starting point of this analysis are shown in Fig. 2. The base case contactor is assumed to have an area of 5000 m^2, and contains a membrane having a CO_2 permeance of 1000 gpu. The feed stream contains 1% CO_2, and the sweep stream is pure nitrogen. The feed and sweep streams are both at atmospheric pressure. The device achieves 80% CO_2 removal from the feed to the sweep gas when the feed and sweep flows are set at 1 m^3(STP)/s.

One concern with high permeance membranes of the type shown in Fig. 2 is that concentration polarization effects may occur in stagnant boundary layers on either side of the membrane. The problem is expected to be most significant on the side in contact with the microporous support layer of the composite membrane. This support creates a stagnant layer which is significantly thicker than the gas boundary layers in the gas channels. The likelihood of concentration gradients forming in the stagnant layer can be estimated by calculating the Peclet number, $J_v\delta/D$, where J_v is the actual gas velocity or volume flux in the layer, δ is the stagnant layer thickness and D is the gas diffusion coefficient in the stagnant layer gas at the stagnant layer pressure [8]. This dimensionless number represents the ratio of the convective transport J_v and the diffusive transport D/δ. When the Peclet number is large ($J_v \geq D/\delta$), the convective flux through the membrane cannot easily be balanced by diffusion in the boundary layer, and concentration gradients form in the boundary layers. When the Peclet number is small ($J_v \leq D/\delta$), convection is easily balanced by diffusion in the boundary layer and significant concentration gradients do not form in the boundary layer. The CO_2 permeance of the membranes shown in Fig. 2 is 1000 gpu (1000×10^{-6} cm^3(STP)/cm^2 s cmHg). Under typical operating conditions of the process in a power plant type of environment (1–2 bar feed, 1 bar permeate and $\sim$10% CO_2 in the feed gas), the volume flux through the membrane is about 1.5×10^{-2} cm^3(STP)/cm^2 s. Assuming the stagnant layer is at atmospheric pressure, the superficial velocity through the microporous support in the layer is 1.5×10^{-2} cm/s. The actual velocity, J_v, will be higher because of the effects of porosity and tortuosity; we will assume here that the actual velocity is about six times higher and equal to 1.0×10^{-1} cm/s. Assuming the microporous support layer that separates the selective membrane layer from the well-mixed counter-flowing gas is 200 μm thick (δ), and taking the gas diffusion coefficient at atmospheric pressure to be $\sim$0.2 cm^2/s, it follows that the permeate-side Peclet number $J_v\delta/D$ is 1×10^{-2}. A Peclet number this small implies that diffusion is