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Application of pilot-scale membrane contactor hybrid system for removal of carbon dioxide from flue gas

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Abstract

A pilot-scale membrane contactor hybrid process was applied to recover CO_2 from the flue gas. Porous polyvinylidenefluoride (PVDF) hollow fiber module was used as membrane contactor and its performance was compared with a conventional packed column. The CO_2 from the flue gas was removed by the gas absorbers using monoethanolamine (MEA) and triethanolamine (TEA) solution as the absorbent. Then, the CO_2 was recovered from the absorbent by the thermal stripping tower with steam and the stripped absorbent was recycled. The membrane contactor increased the available gas–liquid contactor area and mass transfer coefficient, and thus the hybrid process showed a higher CO_2 removal efficiency than the conventional absorption tower. PVDF module with an asymmetric pore structure showed a stable gas–liquid interface, and the CO_2 absorption rate per unit volume of the membrane contactor was 2.7 times higher than that of the packed column. In addition, the membrane contactor hybrid process was successfully operated for 80 h maintaining a greater CO_2 removal efficiency of above 90%. The gas absorption systems were evaluated from the viewpoint of electricity.

Keywords: Membrane contactor; Carbon dioxide; Gas absorption; Hollow fiber; Polyvinylidenefluoride

1. Introduction

The gas absorption process for removing carbon dioxide can be carried out in many reactors such as bubble columns, packed towers, venturi scrubbers and sieve trays. An aqueous solution such as sodium hydroxide, sodium carbonate, monoethanolamine (MEA) or diethanolamine (DEA) is often employed as an absorbent. The main effort in designing and operating the absorber is to maximize the mass transfer rate by producing as much interfacial area as possible. In the case of the packed column, this is made possible by reasonable selection of packing material and uniform distribution of fluids before they enter the packed bed [1,2]. Although the traditional packed bed absorbers have been used in the chemical industry for decades, there are several disadvantages such as flooding at high flow rates, unloading at low flow rates, and channeling and foaming, which lead to difficulties in mass transfer between gas and liquid.

An alternative technology that overcomes these disadvantages and also offers more interfacial area than the conventional approaches is a non-dispersive contactor via a microporous membrane. When a suitable membrane configuration such as a hollow fiber is used, the fluids to be contacted flow on the opposite side of the membrane, and the gas–liquid interface forms at the mouth of each membrane pore. The available contact area remains undisturbed even at a high or low flow rate because the two fluid flows are independent. This porous hollow fiber membrane contactor can be an attractive alternative to packed towers for gas absorption process [3].

In addition, scaling-up is more straightforward with the membrane contactor. Membrane operations usually scale linearly, so that a predictable increase in capacity is achieved simply by adding the membrane modules. Modular design also allows a membrane plant to operate over a wide range

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of capacities. The interfacial area is known and is constant, which allows the performance to be predicted more easily than with the conventional system. With the packed columns the interfacial area per unit volume may be known. However, it is often difficult to determine the loading, that is, what fraction of the available interfacial area is actually used [4].

These advantages have been proved through many researches. Karoor and Sirkar [5] studied the absorption of CO_2 and SO_2 from CO_2/N_2 and SO_2/air mixtures, respectively, into water using a paralleled flow module that employs microporous polypropylene fibers. Kreulen et al. [6] studied the absorption of CO_2 into water/glycerol using polypropylene or polysulfone hollow fibers, and their membrane contactor can be operated stably over a large range of gas and liquid flows without forming foams. Falk-Pesersen and Daninstorm [7] studied on separation of CO_2 from offshore gas turbine exhaust using membrane gas–liquid contactors in both the absorber and the desorber, and optimized the process with respect to size, weight and cost. They reported that compared with conventional absorber columns the use of membrane gas–liquid contactor give a 72% reduction in size and 66% reduction in weight. There are also many researches on CO_2 absorption behavior using various kind of hollow fibers and absorbents [8–10]. Recently, Li and coworkers [11] reported on gas absorptive removal in polyvinylidenefluoride porous asymmetric hollow fiber membrane module. The hollow fiber membrane module is very efficient in the selective removal of trace H_2S to ultra-low concentration even at high gas–liquid flow ratio. In addition, many studies on characterization of the mass transfer behavior for CO_2 absorption into various absorbents have been conducted [12–14]. We have also been studying the polymeric hollow fibers for CO_2 removal from flue gas [15–19]. The successful use of the membrane contactor process over the conventional absorption processes will largely depend on the gas–liquid system and the types of hollow fiber membranes used. In addition to the properties of hollow fibers, operating conditions will also play a major role in the overall membrane contactor performance. However, reports on the combined effect of the conventional process and the properties of hollow fibers on CO_2 removal efficiency are relatively rare. In principle, the conventional absorption process can be replaced by a hollow fiber membrane contactor. Therefore, it is necessary to test the CO_2 absorption capacity using various types of membrane modules combined with the conventional stripper.

In this study, a pilot-scale hybrid process equipped with the membrane contactor instead of the packed tower as the absorber in the conventional process is applied for carbon dioxide removal. The combined process was operated continuously recycling the absorbent through the membrane contactor and the thermal stripper. Porous polyvinylidenefluoride (PVDF) hollow fiber was used as the membrane contactor and the performance of CO_2 absorption was compared with the conventional packed column.

2. Experimental

2.1. Absorption modules

The properties and dimensions of the gas absorption system used in this study are shown in Table 1. Polyvinylidenefluoride (PVDF, Krict Korea) hollow fibers are used as the membrane contactor module, and the hollow fiber is asymmetric as shown in Fig. 1. A cylindrical module of 0.076 m i.d. and 0.52 m length was used as the hollow fiber membrane contactor. For comparison, a packed column was used as the conventional process. The 1/2" PTFE rasching rings were used as the packing material, and the column was 0.094 m i.d. and 2 m length.

2.2. Hybrid process

The experimental apparatus for CO_2 recovery by the hybrid process using membrane contactor are shown in Fig. 2. A gas mixture containing 11 vol.% CO_2 in balance of N_2 was fed into the fiber lumen and was in countercurrent contact with an absorbent solution fed in the shell side of the modules. In the membrane module, the pressure difference of the gas phase and the liquid phase was kept in the range of 2–4 psig by a needle valve in order to form a stable gas–liquid interface. Aqueous solutions of monoethanolamine (MEA, Junsei Chemical Co.) and triethanolamine (TEA, Junsei Chemical Co.) were used as CO_2 absorbent. The carbon dioxide in

Table 1
Dimensions and properties of columns and membrane modules for CO_2 recovery

	Packed column absorber	PVDF module absorber	Stripper
Diameter	0.094 m	0.076 m	0.095 m
Length	2 m	0.52 m	1.56 m
Surface area	372 m^2/m^3	1542 m^2/m^3	–
Volume	0.0138 m^3	0.0024 m^3	–
Hollow fiber membranes	i.d.	830 μm	
	o.d.	1070 μm	
	Pore size	0.03 μm	
	Packing density	0.45	
	Number of fiber	2050	