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CO_2 absorption in a high efficiency silicon nitride mesh contactor

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HIGHLIGHTS

- ▶ A silicon nitride mesh contactor showed excellent performance in CO_2 absorption.
- ▶ The performance was better than other contactors due to the 1 μm mesh thickness.
- ▶ Significant absorption was obtained for contact time less than 1 s.
- ▶ A mathematical model showed good agreement with experimental results.

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ABSTRACT

CO_2 absorption in sodium hydroxide and diethanolamine solutions was investigated in a silicon nitride mesh contactor. Mesh contactors allow two phases to come into direct contact with each other, for the purpose of mass transfer between them without dispersing one phase into the other. The 1 μm thick silicon nitride mesh, containing a high density of uniform 0.5 μm pores, facilitated the stabilization of the gas liquid interface at its pores. Experimental results were obtained for 2 M NaOH or 2 M DEA solutions and 20% vol. CO_2/N_2 inlet concentrations, with a fixed inlet molar ratio CO_2:NaOH of 0.4. Results showed that 23% of the CO_2 contained in the inlet stream was removed within 0.5 s experimental gas residence time. CO_2 removal efficiency was higher when NaOH was used for absorption as compared to DEA. Experiments were also conducted with different mesh/membrane contactors: a PTFE membrane (thickness 20 μm, pore size 0.5–5 μm), a Ni-25 mesh (thickness 25 μm, pore size 25 μm) and a Ni-5 mesh (thickness 5 μm, pore size 5 μm). The silicon nitride mesh demonstrated the best performance primarily due to its small thickness.

1. Introduction

Silicon nitride microsieves [1–9] are manufactured with photolithographic techniques developed in the semiconductor industry. Such micromeshes combine the advantages of minimal mass transfer resistance with high porosity and regular patterned pore structure having at the same time good mechanical strength. They can be used to bring two phases into direct contact with each other, for the purpose of mass transfer between them, without dispersion of one phase into the other. The concept of using micromeshes to bring two phases into contact covers many industrial processes such as extraction, pervaporation, stripping, absorption. Microfabricated meshes are the microengineered analogue of membranes.

Membrane gas absorption is a good alternative to conventional techniques such as packed column absorption. Various investigators have studied CO_2 absorption in membrane contactors. Cussler and co-workers [10] were the first to use microporous polypropylene membranes for H_2S, SO_2 and CO_2 absorption in a NaOH solution, and NH_3 absorption in water. Constantinou and Gavriilidis [11] conducted experimental and theoretical studies using a metallic mesh microreactor to absorb CO_2 in sodium hydroxide solution. Khaisri et al. [12] investigated carbon dioxide absorption into an aqueous solution of monoethanolamine (MEA) using a PTFE membrane contactor. Zhang et al. [13] studied CO_2 absorption in polypropylene (PP) and polyvinylidene fluoride (PVDF) membrane modules using water and aqueous diethanolamine (DEA) solutions as absorbents. Rajabzadeh et al. [14] studied CO_2 absorption in membrane contactors using MEA solutions as the absorbent by using seven different in-house made PVDF and a commercial PTFE hollow fiber membrane with different structures at the outer membrane surface.

In this work, CO_2 absorption in sodium hydroxide and amine solutions was conducted in a high efficiency silicon nitride mesh contactor. Various conditions such as gas and liquid flowrates, type

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Nomenclature

DEA	diethanolamine
D	diffusion coefficient (m^2/s)
F	molar flowrate (mol/s)
k	reaction rate constant (m^3/mols)
m	Henry's constant (ratio of liquid to gas concentrations) (−)
P	pressure (Pa)
r	pore radius (m)
X_{CO_2}	CO_2 removal efficiency
Y	volumetric flowrate (m^3/s)

Greek symbols

γ	surface tension (N/m)
δ	layer thickness (m)
ΔP	pressure difference (Pa)
ε	porosity (-)
θ	contact angle (°)

Subscripts

B	breakthrough
CO_2	carbon dioxide
G	gas phase
i	carbon dioxide, sodium hydroxide
in	inlet
L	liquid phase
NaOH	sodium hydroxide
out	outlet

Superscripts

G	gas phase
L	liquid phase
M	mesh

of flow, liquid chamber height were investigated. The silicon nitride mesh contactor was further compared with other microchannel contactors.

2. Contactor design and experimental conditions

The silicon nitride mesh contactor employed in this work comprised of a silicon microstructured plate with a 1 µm thick layer of silicon nitride, placed between two polycarbonate plates 12 mm thick, containing inlet and outlet ports for the fluids. The 1 µm thick silicon nitride layer was etched using advanced semiconductor processes to form the micromesh. It had well defined pores of 0.5 µm (see Fig. 1) with open area of 20.3%. For the fabrication, a polished monocrystalline silicon wafer 0.675 mm thick was used. It was coated with a 1 µm thick silicon nitride layer by means of chemical vapor deposition. On top of this a photosensitive layer was deposited by spin-coating. A ASML wafer stepper was used for partial exposure of this layer to UV–light. After development, the pattern was transferred into the silicon nitride by reactive ion etching with CHF_3/O_2 plasma. The silicon underneath the perforated silicon nitride mesh was partially removed by anisotropic etching with KOH solution [9]. A top foil made of stainless steel defined a straight liquid channel of 25 µm height, while the gas channel was more complicated and consisted of 675 µm deep chambers within the wafer plus additional 2370 µm chambers from a nickel support; thus the maximum gas channel height was 3210 µm. The gas chamber volume was 1.39 cm^3 based on the total height of 3210 µm, and the liquid chamber volume was 0.01 cm^3. The contactor measured 80 × 64 mm. Two viton gaskets 1 mm thick were placed in 0.75 mm deep grooves in the polycarbonate plates to provide the sealing. The silicon microstructured plate consisted of four blocks and the directions of the fluid streams were perpendicular to the distribution channels. The porous area of the mesh was 42.68 × 9 mm and defined the contact area between the two fluids. Two pin holes were employed in both polycarbonate plates for alignment, while six screws were used for clamping all components together. An HPLC pump (Waters 5100) was used to drive the liquid (2 M NaOH solution or 2 M diethanolamine solution) on the top chamber of the contactor, while the gas 20% vol. CO_2/N_2 was controlled by a mass flow controller (Brooks 5850) and flowed below the mesh. Molar flowrate ratio of CO_2:NaOH was kept at 0.4. Mesh pores were filled by the liquid solutions. The differential pressure between the two phases was controlled by two metering valves (Swagelok) at the outlet of the gas and the liquid phases. The gas phase pressure and liquid phase pressures were measured by pressure sensors (Honeywell; 0–100 kPa) located at the inlets/outlets of the gas and liquid channels. The outlet of the gas phase passed through a liquid trap to avoid any liquid getting into the gas chromatograph (GC) in case of breakthrough of the liquid in the gas phase, and then connected to a GC (Shimadzu GC-14B) for carbon dioxide concentration determination. Experimental data were obtained varying the liquid flowrate in the range 1.28–2.56 ml/min and gas flowrate in the range 160–246 ml/min. These flowrates resulted in residence times of 0.3–0.5 s for the gas (based on gas volume of 1.39 cm^3) and 0.23–0.36 s for the liquid (based on liquid volume of 0.01 cm^3). All the experiments were carried out at room temperature (approximately 20 °C). The CO_2 removal efficiency was calculated from:

$$X_{CO_2} = 1 - \frac{F_{CO_2\,out}}{F_{CO_2\,in}} \quad (1)$$

where F is the molar flowrate of CO_2.

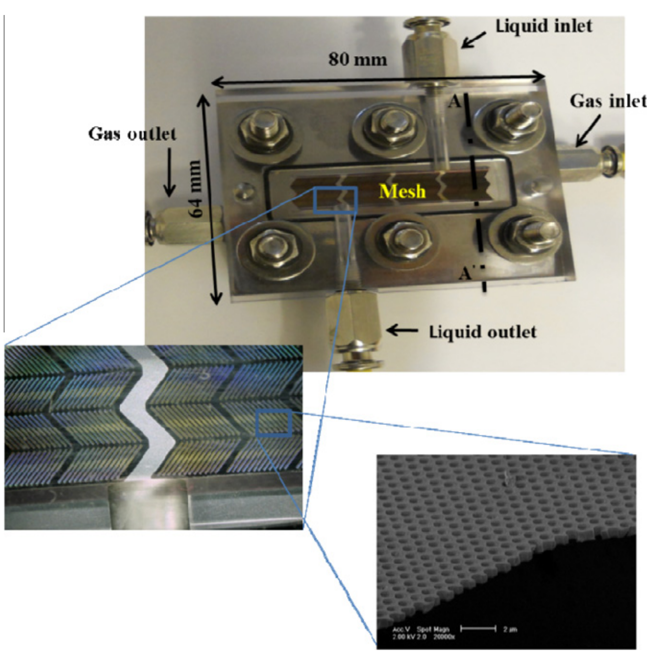

Fig. 1. Picture of the mesh contactor with SEM picture of the silicon nitride mesh.