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Short communication

Separation of N_2/CO_2 mixture using a continuous high-pressure density-driven separator

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ABSTRACT

A mixture of 85% nitrogen and 15% carbon dioxide (N_2/CO_2) is separable at ambient temperature on a continuous basis. A high-pressure, density-driven separator (HDS) was designed and fabricated to explore the process. The effect of the fluid variables, including pressure and mixture flow rate, was assessed on a separation efficiency metric. An important design parameter, the length of the HDS, was also evaluated in the experimental design. Essentially perfect separation is observed over a wide-range of conditions. Separation efficiency is correlated with two dimensionless groups. The first is the Archimedes number. It is a ratio of buoyant force to viscous force. The second dimensionless group is defined in this work. The Espanani number is the ratio of the pressure force to the viscous force. Excellent correlation between separation efficiency and the product of the Archimedes number and the Espanani number is observed. This observation informs both process and equipment design.

1. Introduction

Global warming threatens the environment and humanity in the 21st century. Fig. 1 illustrates temperature anomalies based on data from the National Oceanic and Atmospheric Administration (NOAA). Global temperature has been above the average temperature of the 20th century continually for past 35 years. In 2014, anomaly has reached 0.74 °C. The trend reveals that global temperature will rise 2 °C relative to the 20th century average by the end of this century [1].

Global warming is caused by greenhouse gases such as CO_2, CH_4, NO_x, and SO_x. CO_2 makes up about 80% (by mass) of greenhouse gas emitted by human activities [2]. Therefore, CO_2 emission plays a significant role in global warming. CO_2 concentration in the atmosphere has increased from 280 ppm at the beginning of the industrial revolution, to 380 ppm today [3]. The International Panel on Climate Change (IPCC) predicts it will reach 570 ppm by the end of the century [4].

Combustion of coal, oil, and natural gas emits CO_2. Therefore, separation of CO_2 from flue gas is an important tool to limit global warming. Our work has focused on high-pressure (and relatively low temperature) combustion in supercritical water (SCW). Supercritical water oxidation (SCWO) presents unique opportunities for high-pressure CO_2 separation [5].

Conventional separations of CO_2 in flue gas are based on the molecular, thermodynamic and transport properties of gaseous components in the mixture. The main separation processes include: (1) phase creation (by heat transfer, shaft work or throttling), (2) chemical reaction, (3) absorption, (4) adsorption, (5) permeation through a membrane, and (6) gas hydrate formation [6].

Fig. 2 shows vapor–liquid equilibria (VLE) diagrams of N_2/CO_2 mixture at different temperatures using a Peng Robinson–Wong Sadler-Nonrandom Two Liquid (PR–WS-NRTL) model developed by our group. According to this figure, perfect separation of N_2/CO_2 mixture is impossible via distillation above the triple point temperature of CO_2 ($T > 217$ K). Below the triple point, gaseous CO_2 is directly converted into solid dry ice, which does not favor continuous processing.

If methane is added to the flue gas, its reaction with CO_2 produces syngas (a mixture of H_2 and CO). Thus CO_2 is consumed, but both reactants (CO_2 and CH_4) are thermodynamically stable. The reaction needs a catalyst with considerable activation energy [7].

Amine-based absorption removes acid gases from the flue gas. In this process, the flue gas contacts aqueous solutions of

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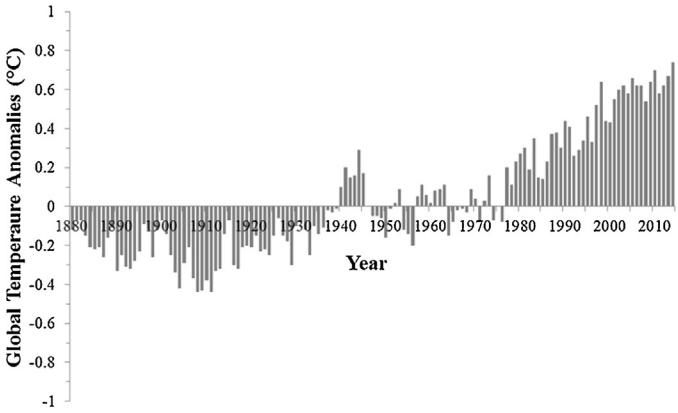

Fig. 1. Annual global anomalies from 1880 to 2014 [1].

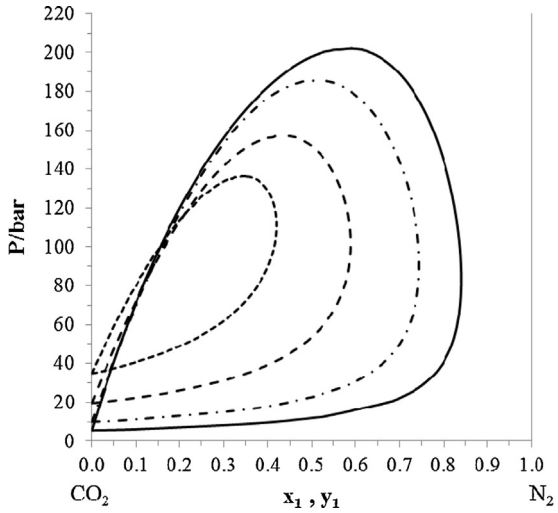

Fig. 2. The diagram of *P-x-y* for the $N_2(1)/CO_2(2)$ mixture at different temperatures. — 218.15 K, ···· 232.85 K, - - - - 253.15 K, and — 273.15 K.

molecules such as monoethanolamine or diethanolamine. The amine functional group takes part in a reversible reaction with CO_2 to form a carbamate. The sorbent is regenerated from the saturated solution by heating and releasing CO_2. Ammonia and alkali salt carbonates can be also used as the solvent in chemical absorption technology. Wide deployment of the technology has not occurred due to high solvent circulation rate, energy intensive regeneration, and corrosion [3,6,8,9].

When the CO_2 partial pressure in a gaseous mixture is greater than 10 bar, a solvent such as methanol and dimethylether polyethylene glycol may be used for physical absorption of CO_2 [10]. Depressurizing the solvent-rich stream regenerates the pure sorbent; therefore the physical absorption technology requires less energy for regeneration than the chemical absorption process. However, a high solvent circulation rate is required because of low absorption of CO_2 by the solvent [6].

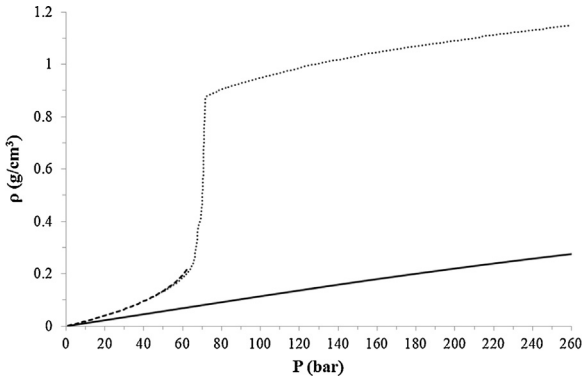

Fig. 3. Densities of pure N_2 and pure CO_2 against pressure at 25 °C. — Density of N_2 at 1–2602 bar calculated by the PR EOS; ···· density of pure CO_2 at 1–63 bar calculated by the PR EOS; — density of pure CO_2 at 45–260 bar measured at our lab.