



# Pre-combustion capture of CO<sub>2</sub> by gas hydrate formation in silica gel pore structure

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## HIGHLIGHTS

- Carbon dioxide (CO<sub>2</sub>) hydrate formation in a fixed-bed type reactor.
- Enhanced CO<sub>2</sub> separation for pre-combustion capture.
- Efficient hydrate-based CO<sub>2</sub> separation process.

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## ABSTRACT

This study presents an efficient hydrate-based CO<sub>2</sub> separation process from a binary mixture consisting of 40.3 mol% CO<sub>2</sub> and balanced H<sub>2</sub>, which is a simulated fuel gas from the integrated gasification combined cycle (IGCC), in the presence of porous silica gel particles containing water. According to our previous work (Y. Seo, S.P. Kang, Enhancing CO<sub>2</sub> separation for pre-combustion capture with hydrate formation in silica gel pore structure, Chem. Eng. J. 161 (2010) 308–312), the cage occupancy of CO<sub>2</sub> in mixed gas hydrates with H<sub>2</sub> is enhanced by the use of a silica gel pore structure containing water. Based on that result, a simulated fuel gas is applied to form gas hydrates, and a richer CO<sub>2</sub> containing gas stream is thereby retrieved through dissociation of the hydrates from a single-stage reactor. Equilibrium dissociation pressures of CO<sub>2</sub> + H<sub>2</sub> gas mixtures were measured with silica gel particles with pore sizes of 25, 100, and 250 nm. The effect of CO<sub>2</sub> concentration on equilibrium dissociation pressures was also investigated at a silica gel pore diameter of 100 nm. The results indicate that when a simulated fuel gas has formed in 100 nm silica gel pores, a gas stream containing more than 96 mol% of CO<sub>2</sub> is achieved by one-stage gas hydrate formation in a silica gel pore structure, which is comparable to the result (88–92) from hydrate formation in bulk water. In addition to demonstrating enhanced distribution of CO<sub>2</sub> in coexisting phases, gas hydrate formation in a fixed-bed type reactor charged with silica gel particles containing water in pores is investigated. The formation of gas hydrates in silica gel pores occurred to a high extent and at a high rate, and the proposed method is thus expected to be a promising CO<sub>2</sub> capture tool for pre-combustion.

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## 1. Introduction

CO<sub>2</sub> emissions from the burning of fossil fuels have been identified as the main contributor to greenhouse gas emissions, which have led to global warming and subsequent climate change. In 2008, 43% of CO<sub>2</sub> emissions from fuel combustion were produced from coal, 37% from oil, and 20% from gas [1]. Coal is fulfilling much of the growing energy demand of developing countries, where energy-intensive industrial production is growing rapidly and is responsible for 80% of power generation. For these reasons,

several techniques have been developed to reduce CO<sub>2</sub> emissions from fossil fuel power plants: oxyfuel combustion, post-combustion CO<sub>2</sub> capture from exhaust flue gas, and pre-combustion CO<sub>2</sub> capture. Oxyfuel combustion is related to improved combustion efficiency, subsequently reducing CO<sub>2</sub> emissions. Post-combustion capture can be achieved in the flue gas stream of power station and is accomplished using amine-based solvents at present. Pre-combustion CO<sub>2</sub> capture involves the removal of CO<sub>2</sub> prior to combustion and is concerned to produce hydrogen (H<sub>2</sub>). H<sub>2</sub> combustion produces no CO<sub>2</sub> emission except water vapor and it is used in a gas turbine or a fuel cell. H<sub>2</sub> is considered an attractive future clean energy source. Even though H<sub>2</sub> production should ultimately be achieved from renewable energy sources, for example solar or wind power, production of H<sub>2</sub> from conventional fossil fuels is

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